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-R.L. CARWIN-

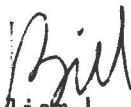
September 11, 1989

TO: Members of the Cold Fusion Panel

Subject: Guidance on the Use of Unpublished Cold Fusion Data

For the final Cold Fusion Panel report, references to unpublished data (e.g., private communications) may be used if required to substantiate an important point. The unpublished data should be considered to be reliable enough to be eventually published, as opposed to very preliminary data. Permission to use unpublished data should be obtained from the author. Referencing preprints will not require obtaining permission from the author.

With regard to obtaining authors' permission to use unpublished data, I suggest that it would be better for the Panel members to do this. Under normal circumstances I would be happy to perform this task, but in this instance your credentials would carry more weight, and furthermore, during the course of your discussions and through a normal give and take you would be in a better position to reach an accommodation and a more productive outcome.

  
William L. Woodard  
Secretary  
Cold Fusion Panel





Government of India  
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# VISION RESULTS IN EXPERIMENTS

*With the Compliments  
from*

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Director

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Fifth International Conference on Emerging  
Nuclear Energy Systems (ICENES V)  
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## COLD FUSION RESULTS IN BARC EXPERIMENTS

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### ABSTRACT

Experiments were initiated at Trombay during the first week of April 1989 to verify the widely reported claims of the occurrence of cold fusion. A large burst of  $\approx 2 \times 10^7$  neutrons was first detected on April 21st with a Pd-Ni electrolytic cell. The neutron counting rate, averaged over a 5 minute interval, was a couple of orders of magnitude larger than that of background count rates. In this experiment the tritium level in the  $D_2O$  electrolyte jumped from the initial stock solution value of 2.6 Bq/ml to  $\approx 5.6 \times 10^4$  Bq/ml, an increase by over four orders of magnitude. The total quantity of tritium generated corresponds to  $\approx 10^{16}$  atoms suggesting a neutron to tritium channel branching ratio of less than  $10^{-8}$  in cold fusion. Significant quantities of neutrons and tritium were also observed to be produced in gas loaded Ti and Pd samples. Autoradiography of  $D_2$  loaded Ti disc targets have shown a number of hot spots indicating uneven distribution of tritium production in the near-surface region. On the whole the Trombay experiments have unequivocally confirmed the occurrence of cold fusion reactions both in Pd and Ti metallic lattices loaded with deuterium.

### 1. $D_2O$ ELECTROLYTIC CELL EXPERIMENTS

Experiments to confirm the cold fusion phenomenon [1,2] were initiated at Trombay during the first week of April 1989. A readily available commercial (Milton-Roy) electrolytic cell [3] with Pd-Ag alloy tubes as cathodes and Ni as anode, originally meant for the generation of  $H_2$  gas was adapted for the electrolysis of 5M NaOD in  $D_2O$ . Both a bank of  $BF_3$  detectors embedded in paraffin and a proton recoil fast neutron detector (NE 102A) were employed to look for possible neutron emission. The first burst of neutrons was detected on 21st April 1989 when the cell current attained 60 amps. Later the current increased to 100 amps and the cell became overheated, resulting in the built-in trip circuit automatically switching off the power. This was followed by a big burst of neutrons approximately

two orders of magnitude larger than background levels during a 5 minute interval. (The  $BF_3$  counter measured 17,758 counts/300s and NE 102A scintillator 25,872 counts/300s as compared to average background values of  $\approx 65$  and 650 respectively). Fig.1 shows plots of the neutron counts data from the two detectors. From the efficiency of the neutron detection system measured using a standard Cf-252 source ( $\approx 2 \times 10^4$  n/s), it was surmised that  $\approx 2 \times 10^7$  neutrons were emitted during that five minute interval.

A number of other  $D_2O$  electrolytic cells with Pd or Ti cathodes of different geometries and designs have also been set up at Trombay, but the current capacities of these cells are lower. Neutron and tritium production has been confirmed in all these cells as well (See Table I).

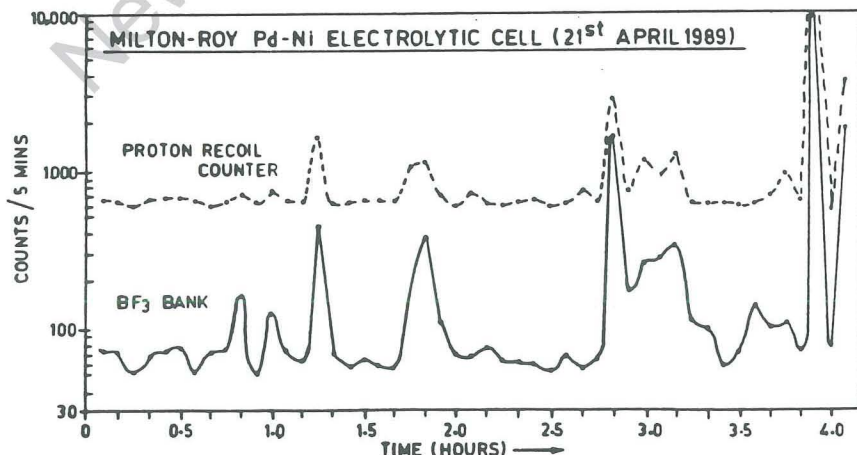


Fig.1. Neutron Counts Data From Two Detectors Using  
Milton-Roy Pd-Ni Electrolytic Cell (21<sup>st</sup> April 1989)

## 2. MEASUREMENT OF TRITIUM LEVELS IN D<sub>2</sub>O ELECTROLYTE

The tritium levels in the D<sub>2</sub>O electrolytes after a few days of continuous operation of the cells were measured by two separate expert groups (belonging to the Isotope and Health Physics Divisions of BARC) who have been engaged in this type of work for over two decades. Well known liquid scintillation counting techniques applicable for low energy beta emitters were used,

taking the following precautions: (a) <sup>40</sup>K free counting vials were employed to minimize background counts. (b) For higher count rate cases, 0.1 to 2 ml of sample was added to the scintillator while for low count rate samples  $\approx$  10 ml was used. In the higher concentration samples pH was reduced by diluting them with double distilled water, in order to minimise chemiluminescence as well as quenching effects. Independently, whenever possible, these values were cross-checked with results obtained after "chemiluminescence cooling". (c) Commercially available Instagel scintillation cocktail was preferred over Dioxane as a solvent to minimize chemiluminescence interference effects.

Prior to the commencement of electrolysis samples of the initial electrolyte were saved and counted along with each sample drawn during the course of the experiment. To compute the excess tritium produced in each run, the following points were taken into account: (a) Initial tritium concentration in the stock D<sub>2</sub>O; (b) To be on conservative side the tritium that is carried away by the D<sub>2</sub> gas stream during electrolysis was neglected even though it is known that above

20°C the (T/D) ratio in the gas stream is comparable to that in the liquid phase and (c) Dilution effects due to periodic make-up of D<sub>2</sub>O. Table I presents the tritium yields measured in some of the electrolytic cell experiments conducted at Trombay.

## 3. MULTIPLICITY SPECTRUM OF NEUTRON EMISSION

It is of interest to establish the multiplicity spectrum of neutron emission i.e. whether the neutrons are emitted individually (one at a time) or in sharp bursts of 10 or more at a time. The number of neutron pulses issuing from a bank of BF<sub>3</sub> counters (embedded in paraffin) monitoring the Milton-Roy electrolytic cell was totalled over 10 ms sampling intervals and stored in a personal computer. There were 1000 such sampling intervals during a real time of 5 minutes. Data was simultaneously also recorded from a paraffin

encased bank of <sup>3</sup>He neutron detectors placed at a distance of  $\approx$  1.5 m from the cell, serving as a background monitor. The data accumulated during periods of significant neutron emission were statistically analysed to yield the probability distribution of neutron counts. It may be readily inferred from Table II which presents one such result, that neutron emission essentially obeys Poisson distribution. However it is also found that occasionally more than 10 neutron counts are registered in a single 10 ms interval. The background monitor has never yielded such high multiplicity events even over a one week period of continuous monitoring. This points to the occurrence of nuclear events wherein 100 or more

TABLE I: TRITIUM PRODUCTION IN ELECTROLYTIC COLD FUSION EXPERIMENTS

Sl No	DIVISION /GROUP	CATHODE MATERIAL /SHAPE	ELECTROLYTE	VOL. OF D <sub>2</sub> O SOLN.	MAX. CELL CURRENT (Amps)	TRITIUM LEVELS (Bq/ ml)		TRITIUM PRODUCTION (Bq) Atoms	
						INITIAL	FINAL	(10 <sup>6</sup> )	(10 <sup>14</sup> )
1.	HWD/NtPD	Pd-Ag Tubes (M-R Cell)	5M NaOD in D <sub>2</sub> O	250 ml	100	2.6	55.6	13.9	80
2.	"	"	"	"	"	10.0	4.4	1.1	6
3.	HWD/DD/NtPD	Pd Sheets	"	1000 ml	65	2.0	7.0	7.0	40
4.	"	Ti Rod	"	135 ml	40	2.0	1.8	0.24	1.3
5.	ACD-(i)	Pd Cylinder	0.1 M LiOD in D <sub>2</sub> O	45 ml	1-2	31.3	16.6	0.75	4
6.	ACD-(ii)	Pd Ring	"	65 ml	1-3	18.1	8.8	0.57	3

neutrons are emitted in a single sharp burst, probably under a  $100\mu\text{s}$  in duration. Such occasional neutron "flashes" are also reported to have been observed by Menlove et al at LANL /4/.

#### 4. GAS LOADED TITANIUM TARGET EXPERIMENTS

Following reports of neutron emission having been detected by the Frascati group /5/ with pressurised  $\text{D}_2$  gas loaded Ti shavings, two variants of this experiment were carried out at Trombay. One group (Chemistry Division) followed the Frascati procedure with  $\approx 20$  g of cut Ti pieces and  $\text{D}_2$  gas pressures increasing upto 50 bars. The temperature was cycled between 77 K and room temperature. The neutron detection system comprised of 24  $^3\text{He}$  counters arranged in a well like array and having a counting efficiency of  $\approx 10\%$ . The first neutron emission from this set up occurred on 3rd June when the temperature increased to ambient value from 77 K with simultaneous evacuation. The neutron count rate reached a peak value of 3900/40s as compared to initial background levels of 60 per 40s. The neutron emission phase lasted for about 30 minutes. The same charge was then subjected to temperature cycling between 77 K and 300 K leading to emission of  $\approx 6.5 \times 10^6$  neutrons in all

over a 7 hour duration. A similar neutron output lasting for  $\approx 2$  hours was recorded subsequently when the sample temperature was raised gradually with simultaneous evacuation. Fig.2. depicts the neutron yield variation measured during this run.

In a second variation of the gas loading experiment, small machined targets (discs, cones, cylinders etc) of Ti metal (mass between 0.2 to 1 g) were individually loaded with  $\text{D}_2$  gas by heating them to  $900^\circ\text{C}$  in  $\text{D}_2$  atmosphere at 1 Torr pressure and then switching off the power to the induction coil.  $\text{D}_2$  gas was absorbed by the Ti target in the course of a minute or so during its cooling. The quantity of  $\text{D}_2$  absorbed could be measured from the observed pressure drop. This corresponded to  $\approx 10^{10}$  molecules of  $\text{D}_2$  gas, indicating a gross (D/Ti) ratio of hardly 0.001. However it is believed that most of the absorbed  $\text{D}_2$  gas is accumulated in the near surface region. Some targets did show statistically significant excess-over-background neutron counts during the one minute duration of gas absorption. A disc shaped Ti button loaded on Friday 16th June began emitting neutrons on its own, almost 50 hours after loading. It produced  $\approx 10^6$  neutrons over a 85 minute active phase. The background neutron counter did not show any increase in counts during this time.

TABLE II: MULTIPLICITY DISTRIBUTION OF NEUTRON COUNTS IN 10 ms INTERVALS

(Milton-Roy Electrolytic Cell: Friday 16th June 1989)

Time (Hrs)	BF <sub>3</sub> Counter Bank (Signal)															<sup>3</sup> He Counter Bank (Background)		
	1*	2*	3*	4*	5*	6*	7*	8*	9*	10*	11*	12*	13*	14*	15*	1*	2*	3*
18.55	124	21	4	1	—	—	—	—	—	—	—	—	—	—	—	2	1	—
19.00	54	9	1	—	—	—	—	—	—	—	—	—	—	—	—	1	—	—
19.05	335	54	7	2	1	—	—	—	—	—	—	—	—	—	—	4	—	—
19.10	320	82	10	—	—	—	—	—	—	—	—	—	—	—	—	5	—	—
19.15	243	13	4	—	1	—	—	—	—	—	—	—	—	—	—	5	—	—
19.20	315	35	3	1	—	—	—	—	—	—	—	—	—	—	—	4	—	—
19.25	295	24	—	1	—	—	—	—	—	—	—	—	—	—	—	5	—	—
19.30	492	51	3	2	—	—	—	—	—	—	—	—	—	—	—	4	—	—
19.35	447	42	2	1	—	—	—	—	—	1	—	1	—	—	—	9	—	—
19.40	104	13	4	—	—	1	—	—	—	—	—	—	—	—	—	5	—	—
19.45	355	49	1	1	—	1	—	—	—	—	—	—	—	—	—	33	1	—
19.50	395	99	16	2	—	—	—	—	1	—	—	—	—	—	—	22	3	—
19.55	55	24	7	33	2	1	1	1	1	1	2	2	1	—	5	6	2	—

(\*) Starred numbers represent the multiplicity of counts obtained in a single 10 ms interval. The respective frequency of occurrence (per 1000 gated intervals) is given in the corresponding column below.



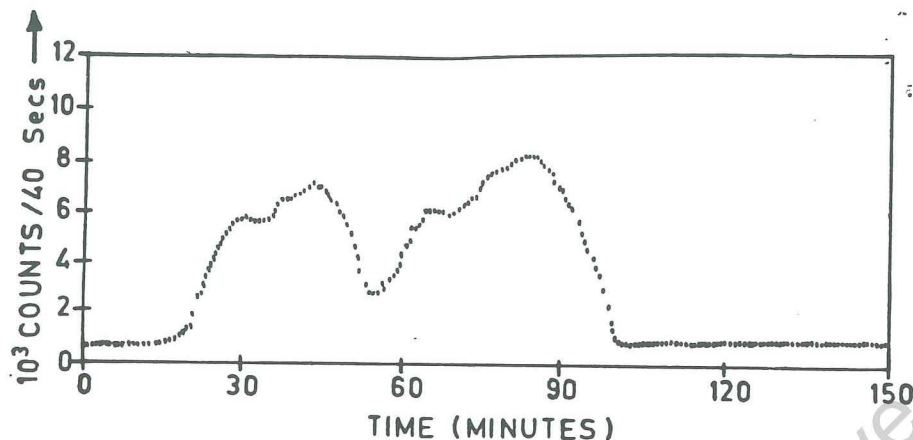


Fig.2. Neutron-Yield Variation During Desorption Mode: D<sub>2</sub>O Gas Loaded  
Titanium-Experiments (7<sup>th</sup> June 1989)

##### 5. AUTORADIOGRAPHY OF DEUTERATED TITANIUM TARGETS

Since tritium which emits betas of 18 Kev (end point energy) has been found to be the primary product of cold fusion, it may be expected that autoradiography of deuterated Ti targets may give very useful information in the form of space resolved images. Deuterated Ti targets in which cold fusion reactions were suspected to have occurred were placed over a standard medical X-ray film and exposed overnight. On developing, the radiographs of deuterated Ti discs showed about a dozen intense spots randomly distributed within the disc boundary, besides a large number of smaller spots, especially all along the periphery forming a neat ring of dots (see Fig.3). Repeated measurements with the same disc target with exposure times varying from 10 to 40 hours gave almost identical patterns of spots, indicating that the tritium containing regions were well entrenched in the face of the titanium lattice. Spectral analysis of the X-ray emissions from such targets using a Si(Li) detector clearly showed the characteristic K<sub>α</sub>(4.51 Kev) and K<sub>β</sub>(4.931 Kev) peaks of titanium excited by the betas from tritium decay.



Fig.3. Typical Autoradiograph of a Deuterated Ti Disc Showing Regions of Tritium Concentration

## 6. SUMMARY AND CONCLUSIONS

Investigations of cold fusion phenomena carried out at Trombay during the first three months of the "cold fusion era", namely April to June 1989, have positively confirmed the occurrence of (d-d) fusion reactions in both electrolytic and gas loaded Pd and Ti metal lattices at ambient temperatures. Neutron emission has been observed even when the current to the electrolytic cell is switched off or in case of gas loaded Ti targets when no externally induced perturbation such as heating/ cooling/ evacuation etc is effected. The main findings of the Trombay investigations to date may be summarised as under:

(a) Tritium is the primary product of cold fusion reactions, notwithstanding the fact that the tritium, if any, entrapped inside the palladium electrodes has yet to be quantitatively assessed. Thus cold fusion may be characterised as being essentially "aneutronic" with a neutron to tritium channel branching ratio of less than  $10^{-8}$ .

(b) Neutron emission both from electrolytically loaded Pd and gas loaded Ti is basically Poisson in nature i.e. the neutrons are emitted one at a time. However it is not clear whether the neutrons are generated in the (d-d) fusion reaction itself or whether it is produced in a secondary reaction involving the energetic protons or tritons. In this context it would be of interest to look for the possible presence of 14 Mev neutrons in cold fusion experiments.

(c) Occasionally nuclear events do appear to take place wherein over a 100 neutrons are generated in a single sharp burst. Viewed in the light of the branching ratio estimate of  $10^{-8}$  noted above, this leads to the intriguing conclusion that a chain reaction involving as much as  $10^{10}$  fusion reactions occurs within a time span of a 100  $\mu$ s.

(d) Autoradiography of gas loaded Ti targets demonstrates in a simple and elegant manner not only the occurrence of cold fusion, but also the production of tritium. The estimated tritium to deuterium isotopic ratio in these targets is several orders of magnitude higher than in the initial stock  $D_2O$  and as such cannot be explained away on the basis of preferential absorption of tritium by the titanium as may be suspected. The existence of highly localised regions (hot spots) on the target surface wherein tritium is concentrated as well as the occurrence of spots all along the periphery of the disc, points to the important role of lattice defect-sites in the absorption process or in the accumulation of tritium following migration after its formation, at least in titanium.

The very high probability for the tritium branch in cold (d-d) fusion reactions would indicate processes of neutron transfer across the potential barrier as postulated by Oppenheimer over half a century ago /6/ and elaborated on more recently by Rand McNally /7/. If neutron transfer as envisaged by these authors does take place so easily, it may have many implications for the future of nuclear technology, for the deuterium nuclide might very well do the work which free neutrons do in present day fission reactors. In the context of the emerging energy production scenario, aneutronic fusion reactions such as this may give rise to new fusion technologies providing a cleaner energy source for the twenty first century.

## ACKNOWLEDGEMENT

The work described in this paper has been carried out by many groups of scientists at BARC, Trombay. The details of the various experiments are under publication as a consolidated BARC report. I am grateful to many of my scientific colleagues for the illuminating discussions on various aspects of the work presented here. I wish to particularly thank Dr. M. Srinivasan for his help in the preparation of this paper.

## REFERENCES

- 1 Fleishmann, M and Pons, S., Electrochemically Induced Nuclear Fusion of Deuterium, J. Electroanal. Chem. 261, 301-308 (1989)
- 2 Jones S.E. et al, Observation of Cold Nuclear Fusion in Condensed Matter, Nature 338, 737-740 (1989)
- 3 ELHYGEN Hydrogen Generator (Mark V), Manufactured by Milton-Roy Company, Shannon Industrial Estate, Clare, Ireland
- 4 Report on the Workshop on Cold Fusion held at Santa Fe, New Mexico, USA during May 1989, Science News, June 3rd, 1989
- 5 De Nino A et al, Evidence of Emission of Neutrons From a Titanium - Deuterium System, Submitted for Publication to Europhysics Letters (1989)
- 6 Oppenheimer, J.R. and Philips, M., Note on the Transmutation Function for Deuterons, Phys. Rev. 48, 500 - 502 (1935)
- 7 Rand McNally, Jr. J, Cold Fusion Prospects, Bulletin of APS, Baltimore Meeting, April 1983.

# Cold Fusion: A Hypothesis

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ABSTRACT: It is suggested that the evidence for the putative phenomenon of cold fusion is valid, but that the effect is not dominated by a DD reaction.

On March 23, 1989, in a news conference at the University of Utah, the electrochemist B. S. Pons (also speaking for his colleague M. Fleischmann) claimed<sup>1</sup> that nuclear energy, in the form of heat, was liberated by a room temperature "table-top" apparatus that used a palladium (Pd) cathode to electrolyze heavy water (D<sub>2</sub>O). In a paper submitted for publication a few days earlier,<sup>2</sup> the two chemists had written that "the bulk of the energy release is due to a hitherto unknown nuclear process or processes (presumably .... due to clusters of deuterons)." The immediate -- and thereafter unrelenting -- reaction of the hot fusion community was disbelief, based on the absence, at the expected intensities, of the customary signs of a DD reaction, such as neutrons ( $d + d \rightarrow n + {}^3\text{He}$ ) and high energy  $\gamma$ -rays ( $d + d \rightarrow \gamma + {}^4\text{He}$ ).

The hypothesis that I now advance has the following ingredients:

- (1) The claim of Pons and Fleischmann to have realized cold fusion is valid.
- (2) But, this cold fusion process is not powered by a DD reaction. Rather, it is an HD reaction, which feeds on the small contamination of D<sub>2</sub>O by H<sub>2</sub>O.
- (3) The HD reaction  $p + d \rightarrow {}^3\text{He}$  does not have an accompanying  $\gamma$ -ray; the excess energy is taken up by the metallic lattice of Pd alloyed with D. (Others have mentioned the possible importance of an HD reaction, but without reference to the lattice, and with no claim for its dominance over DD reactions.)
- (4) The coupling with the Pd-D lattice that rapidly siphons off nuclear energy, as it becomes available, had previously acted to suppress the Coulomb repulsion between p and d, and, indeed, to overcome it with an energy of attraction that significantly ameliorates the effect of Coulomb barrier penetration.
- (5) The asymmetry of the pd situation, compared with the symmetry of dd, enhances the HD reaction over DD reactions.

In this view, the dominant fusion reaction produces neither neutrons nor high energy  $\gamma$ -rays. Of course, the same could be said of a purely chemical reaction. But the specific fusion hypothesis outlined here invites a critical



experimental test, one to which neither a DD nuclear reaction nor an (unknown) chemical reaction would appear to be sensitive: Compare the heat outputs of cells that are similarly prepared in all respects but the varied small admixtures of  $H_2O$  in the  $D_2O$ . This may be an overly idealized proposal, however. The short, bloody, history of cold fusion indicates that 'similarly prepared' is not a trivial condition. An alternative, and apparently simpler, procedure is to add a small percentage of  $H_2O$  to a functioning cell, but here one must not allow the long term effects to be obscured by transient phenomena. Finally, concerning the oft repeated demand for a control experiment using  $H_2O$ , one should note the possibility of a converse effect of the HD reaction: Through the natural presence of  $D_2O$  in ordinary water, such a control experiment might produce an otherwise puzzling amount of heat.

The details of the lattice coupling mechanism will be presented elsewhere.

I am grateful for helpful exchanges with R. Finkelstein, UCLA, and J. Simons, University of Utah.

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1. New York Times (National Edition), March 24, 1989, p. A 16.

2 M. Fleischmann and S. Pons, "Electrochemically Induced Nuclear Fusion of Deuterium, As Submitted March 20, 1989, to Journal of Electroanalytical Chemistry.

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September 13, 1989  
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Washington, DC 20585

Dear Bill,

Enclosed is a very interesting response from P.K. Iyengar to my letter of 09/07/89. Please distribute this to the Cold Fusion Panel.

I do not believe the argument of the second paragraph.

Sincerely yours,



Richard L. Garwin

Encl:

09/13/89 TELEX P.K. Iyengar to RLG. (091389.PK1)

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-R.L. GARWIN-

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FOR: DR. RICHARD L GARWIN IBM RESEARCH DIVISION THOMAS J WATSON  
RESEARCH CENTER NY 10598

FROM: DR P K IYENGAR DIRECTOR B A R C BOMBAY INDIA  
(FAX NO. 2048476)

DEAR DR. GARWIN:

THANK YOU FOR YOUR TELEX OF SEPTEMBER 7, 1989. SINCE THE PRESENTATION OF MY PAPER AT THE KARLSRUHE MEETING WE HAVE CONTINUED TO EXPERIMENT WITH PALLADIUM AND TITANIUM, BOTH BY ELECTROLYSIS AND GAS ABSORPTION. WE CONSISTENTLY SEE TRITIUM AS WELL AS NEUTRON PRODUCTION IN BURSTS. THE SAME CELLS WITH HIGHER CURRENT HAVE PRODUCED NEUTRON BURSTS AS WELL AS TRITIUM BURSTS. BURSTING NATURE OF TRITIUM PRODUCTION ALONG WITH NEUTRONS HAS BEEN OBSERVED BY US RECENTLY WITH CELL 1 IN TABLE 1 BY USING AN ON-LINE TRITIUM MONITOR ALONG WITH A NEUTRON DETECTOR. THIS TIME WE USED GM COUNTERS WITH SILVER CATHODES TO DETECT NEUTRONS BY ACTIVATION. NEW CELLS HAVE ALSO BEEN SUCCESSFUL. THE AUTORADIOGRAPHIC TECHNIQUE HAS BEEN TRIED OUT WITH GAS LOADED PALLADIUM FOILS AND HAS SHOWN SIMILAR RESULTS AS DESCRIBED IN MY PAPER EXCEPT THAT WE DO NOT SEE SPOTS IN THE CASE OF PALLADIUM.

AS REGARDS YOUR QUERY REGARDING BRANCHING RATIO, IT IS TRUE THAT ENERGETIC TRITONS SHOULD PRODUCE 14 MEV NEUTRONS IN THE PALLADIUM MATRIX. THE BRANCHING RATION OF  $10\pm 8$  QUOTED IN OUR PAPER WAS DEDUCED USING ONLY THE NEUTRONS OBSERVED DURING BURSTS. HOWEVER, IF ADDITIONAL AMOUNT OF NEUTRONS POSSIBLY PRODUCED BETWEEN BURSTS BUT SUBMERGED IN THE BACKGROUND IS TAKEN INTO CONSIDERATION ALONG WITH THE LOWER EFFICIENCY FOR DETECTION OF 14 MEV NEUTRONS, THE NEUTRONS TO TRITIUM BRANCHING RATION WOULD BECOME SOMEWHAT HIGHER, PERHAPS  $10\pm 6$  OR  $10\pm 5$ . ALTHOUGH THE INCONSISTENCY MENTIONED BY YOU WOULD DISAPPEAR IT WOULD IMPLY THAT BULK OF THE NEUTRON PRODUCED WOULD BE OF 14 MEV ENERGY. WE ARE, THEREFORE, TRYING TO MEASURE THE SPECTRUM OF NEUTRON EMISSION CAREFULLY. HOWEVER, IT MAY ALSO TURN OUT THAT THE INTEGRATED PROBABILITY OF A(TD) REACTION CAUSED BY TRITONS RELEASED WITHIN THE LATTICE MAY BE DIFFERENT COMPARED TO MEASURED BEAM TARGET VALUES. IN ANY CASE THE ABOVE ARGUMENTS WILL LEAD TO THE CONCLUSION THAT THE TRUE BRANCHING RATIO IN THE (DD) FUSION IS SMALLER THAN  $10\pm 8$  FOR NEUTRON PRODUCTION CONFIRMING THAT COLD FUSION IS ESSENTIALLY ANEUTRONIC IN NATURE.

AS REGARDS THE CONSOLIDATED BARC REPORT, IT IS STILL UNDER PREPARATION EVENTHOUGH SEVERAL PARTS OF IT ARE READY. I SHALL MAIL IT TO YOU THE MOMENT THIS IS COMPLETE, HOPEFULLY WITHIN A FEW WEEKS.

LET ME ALSO ENDORSE YOUR OBSERVATION "SCIENCE MARCHES ON", IN SPITE OF GOOD AND BAD REPORTS.

WITH BEST WISHES,

SINCERELY YOURS

P.K.IYENGAR

OVER PL ACK

IBMRESRCH YKHG UD  
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MMMM

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To: Dr. William L. Woodard (Via FAX to 9 (202) 586-3119)

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September 12, 1989  
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Dr. William L. Woodard  
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to the  
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Dear Bill,

In preparing to write my section on neutrons in the "fusion products" part of the report, I have just spoken with Dr. Marcello Martone of Frascati. You remember that I visited Dr. Martone in his laboratory 06/09/89, to follow-up on the beautiful results of April on neutron bursts, and on the continuous neutron emission over 16 hours that were presented in the Frascati preprint by Scaramuzzi and co-workers. This work was also presented at the Santa Fe conference.

Although Dr. Martone returned from his vacation only yesterday, and so may not be up on the very latest results, he tells me that they have had no neutrons from their "dry fusion" experiments since those first results in April.

Martone says also that they shut off their electrochemical cells in July, having had no positive results from them either.

Martone had not heard of the work at BARC. I promised to tell him what I learned from my Telex communication with Dr. Iyengar at BARC, and Martone promised to send me by FAX any positive results from dry fusion from Frascati.

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ARGONNE NATIONAL LABORATORY

Date: Sept. 12, 1989

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**Nature of Short Range Interaction Between Deuterium Atoms in Pd**

**Feng Liu, B. K. Rao, S. N. Khanna, and P. Jena**

**Physics Department**

**Virginia Commonwealth University**

**Richmond, Virginia 23284-2000**

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### Abstract

The likelihood that two deuterium atoms in Pd could approach each other close enough to fuse under ambient conditions has been examined by calculating energy costs self-consistently as a function of separation distance,  $R$ , using both Hartree-Fock and local density approximations. A minimum in the potential energy curve exists at  $R \sim 1.1 \text{ \AA}$  along the (100) direction, which is larger than the covalent bond length of  $0.74 \text{ \AA}$  in the free molecule. For very short distances,  $R \lesssim 0.2 \text{ \AA}$ , the interaction between the deuterium atoms is virtually identical to that in free space, thus making conventional fusion unlikely.

The announcement by Fleischmann and Pons<sup>1</sup> and, independently, by Jones et al.<sup>2</sup> that two deuterium atoms inside a Pd matrix could fuse has created a level of excitement in the scientific, as well as the media world, not seen since the days of high  $T_c$  superconductivity. While little scientific information can be extracted from publications in newspapers and reports in radio and television, the following picture seems to be apparent. If fusion of deuteriums in Pd is true, then the basic interaction between the deuterium atoms at close distances must be qualitatively different from that in the free space. For example, one hopes that the conduction electrons in Pd would act much like negative muons in bringing the two deuterium atoms close enough to induce fusion. Rafelski et al.<sup>4</sup> have examined the role of three different parameters; the effective mass,  $m^*/m$  and the effective charge,  $e^*/e$  of the host conduction electrons and the separation distance  $R$ , between the D-atoms, on the fusion rate. They point out that for the free  $D_2$  molecule with the equilibrium separation distance of  $0.74\text{\AA}$  the fusion rate is of the order of  $10^{-74}/s$ . However, the fusion rate depends very sensitively on the three parameters mentioned above. For example, using  $m^*/m=2$ ,  $e^*/e = 1.5$  and  $R = 0.5a_0$ , the fusion rate is estimated<sup>4</sup> to increase to an observable range of  $10^{-20}/s$ .

The basic question is then an obvious one. How close can the D-atoms come together in Pd and what are the corresponding



energy costs? Is the interaction between D-atoms in Pd basically different from that in other metals? If not, could "fusion" be achieved in a less expensive metal?

In this letter we examine some of these issues. We calculate the energies of a pair of D-atoms separated by a distance  $R(0.05\text{\AA} < R < 2\text{\AA})$  in Pd, Li, and Al metals. Both Pd and Al were modelled by a cluster of six metal atoms forming an octahedron (see Fig. 1) and Li by a cluster of ten atoms. The energies of the ambient clusters as well as the clusters with two D-atoms at varying separation were calculated self-consistently using the linear-combination of atomic orbitals - molecular orbital (LCAO-MO) technique.

We start with a set of molecular orbitals given by

$$\psi_i(\vec{r}) = \sum_{\mu} \sum_k C_{\mu i}^k \phi_{\mu}^k(\vec{r} - \vec{R}_{\mu}) \quad (1)$$

where  $R_{\mu}$  is the site of the  $\mu$ th atom and  $\phi_{\mu}^k$ 's are the atomic wave functions corresponding to the orbital  $k$  centered at  $R_{\mu}$ . We treat the D-atoms within the Born-Oppenheimer approximation. Consequently, both deuterium and hydrogen atoms are indistinguishable from each other as far as energetics is concerned. The coefficients  $C_{\mu i}^k$  are evaluated self-consistently by solving the variational equation,

$$H\psi_i = E_i\psi_i. \quad (2)$$

The electronic Hamiltonian is expressed in atomic units by,

$$H = -\frac{1}{2}\nabla^2 + V_{es}(\vec{r}) + V_{ex}(\vec{r}) + V_{corr}(\vec{r}) \quad (3)$$

The first two terms are the kinetic and electrostatic term that can be evaluated without any approximation. The last two terms, exchange and correlation, are difficult to evaluate and it is conventional in condensed matter physics to express these by the local density approximation (LDA) within the framework of the density functional theory. However, for the problem at hand where the two D-atoms could approach as close as  $0.05\text{\AA}$ , one would have to wonder about the validity of the local density approximation. An alternate method, and certainly a more rigorous one, is to calculate the energetics using Hartree-Fock theory for the exchange and correlation through configuration interaction. Unfortunately, the latter technique is computer intensive and cannot easily be used to study PdD systems involving many Pd atoms.

To compute the energetics of  $\text{Pd}_6\text{D}_2$  clusters we have used the discrete variational method<sup>5</sup> (DVM) and the local density approximation to the exchange-correlation potential. For Pd, we have frozen the inner core-orbitals 1s, 2s, 2p, 3s, 3p and 3d. The outer orbitals 4s, 4p, 4d, 5s and 5p were included in the variational procedure. For D-atom, we used the 1s, 2s, and 2p

basis sets. The energy integration employed up to 8000 points for Pd and 3000 points for D. Our calculation yields the bond length and binding energy of PdH to be  $1.53\text{\AA}$  and 2.74 eV respectively. A relativistic calculation by Balasubramanian et al.<sup>6</sup> involving quantum chemical technique yields the bond length and binding energy of PdH to be  $1.53\text{\AA}$  and 2.21 eV respectively. The calculated bond lengths are in excellent agreement with experimental value<sup>7</sup> of  $1.53\text{\AA}$ . The experimental value<sup>7</sup> for the binding energy is 3.3 eV. However, as Balasubramanian et al.<sup>6</sup> have pointed out, the experimental value is quite uncertain and the expected value would lie<sup>6</sup> between 2.45 eV-2.75 eV.

For  $\text{Li}_{10}\text{H}_2$  and  $\text{Al}_6\text{H}_2$  clusters we used the Hartree-Fock theory with Gaussian basis sets. The details of our work in Li and Al metals will be published later. In this letter, we concentrate on our results in Pd and discuss the results in Li and Al only as a reference. To assess the reliability of LDA results in accounting for the D-D interaction at short distances, we first compare in Fig. 2, the LDA energies (measured with respect to dissociated atoms) as a function of distance in free space with that using the Hartree-Fock (HF) method, namely

$$\Delta E_0(R) = E_R(D_2) - 2E_0(D) \quad (4)$$

$E_R(D_2)$  is the energy of the two D-atoms a distance R apart and  $E_0(D)$  is the D-atom energy. Note that at all distances, the two

energies compare very well. The LDA energy is slightly lower than the HF result. This is expected since correlation always lowers the energy. Both HF and LDA energies yield the correct  $D_2$  bond length of  $0.74\text{\AA}$ . The binding energies of  $D_2$  using LDA and HF with correlation via Möller-Plesset<sup>8</sup> scheme are respectively 4.54 eV and 4.57 eV. These compare well with the experimental value of 4.45 eV.

The results in Fig. 2 as well as those for the dimers give us confidence that the energetics of D-atoms in Pd using the LDA would be reliable. Using the DVM code<sup>5</sup>, we compute,

$$\Delta E(R) = E_R(\text{Pd}_6\text{D}_2) - E_0(\text{Pd}_6) - 2E_0(\text{D}) \quad (5)$$

where  $E_R(\text{Pd}_6\text{D}_2)$  is the energy of the  $\text{Pd}_6\text{D}_2$  cluster with the D-atoms separated by a distance  $R$  along the (100) as well as (111) directions. The Pd-atoms occupy the lattice sites as they would in a Pd metal (see Fig. 1).  $E_0(\text{Pd}_6)$  is the energy of the isolated  $\text{Pd}_6$  cluster.  $\Delta E(R)$  is, therefore, the energy cost in bringing the two D-atoms together to within a distance  $R$  in Pd.

In Fig. 3 we compare  $\Delta E(R)$  in Eq. (5) with that in Eq. (4). Two salient features are to be noted. First for  $R \lesssim 0.2\text{\AA}$ , the curves  $\Delta E(R)$  and  $\Delta E_0(R)$  are virtually identical<sup>9</sup> suggesting that the electrons of Pd atoms do not play any significant role in bringing the D-atoms closer. Thus, if for a reasonable fusion



rate one would require the D-D distance to be about  $0.3\text{\AA}$ , hydrides are not the place to look for assistance. There are, however, other features in  $\Delta E(R)$  for  $R > 0.7\text{\AA}$  that are quite interesting. To show this more clearly, we amplify this portion in Fig. 4. First, note that  $\Delta E(R)$  has a minimum along the (100) direction at  $R \sim 1.1\text{\AA}$  while no such feature exists along (111) direction. This minimum arises since the motion of D-atoms along (100) is hindered by the Pd-atoms on the face center. In  $\text{Pd}_6\text{D}_2$  cluster, there are no Pd-atoms to block the motion of D-atoms along the (111) direction. Consequently the potential is repulsive at all ranges shown in Fig. 4. We have repeated the energy calculations in a  $\text{Pd}_8\text{D}_2$  cluster by capping the two corner sites along the (111) direction. As expected we do find a minimum in the energy. However, this occurs for a much larger  $R > 3.4\text{\AA}$ . Secondly, the energies in Fig. 4 along (111) are consistently lower than those along (100) for  $R > 0.7\text{\AA}$ . For smaller D-D separation, the two energies are nearly the same. This is as expected, since for small distances, the interactions between the D-atoms dominate over those between Pd and D. Since the energy at the minimum along (100) direction is higher than that along (111), one could conclude that the  $\text{D}_2$  pairing in Pd may not be possible. It is worth pointing out that no  $\text{D}_2$  pairing in Pd has been observed in NMR experiments<sup>10</sup>.

Recently, Mintmire et. al.<sup>11</sup> have calculated the energetics of  $\text{D}_2$  in Pd using a cluster approach similar to that described

here. These authors have used the Gaussian basis functions, instead of numerical ones as used here, and the local density approximation. They found the energies along (111) direction to be repulsive, in agreement with our results. Dunlap and coworkers<sup>12</sup> have also calculated the energies of D<sub>2</sub> pair along the (100) direction in a Pd<sub>6</sub>D<sub>2</sub> cluster. They found a minimum in the energy curve along (100), for  $R \sim 1\text{\AA}$ . This minimum, as in our calculation, is higher than the energies along (111) direction. Note that the minimum obtained by Dunlap at  $R \sim 1\text{\AA}$  is somewhat different from our result of  $\sim 1.1\text{\AA}$ . Comparison of our results with that of Dunlap and coworkers<sup>11,12</sup> clearly reveal that the D-atoms cannot approach closer than  $0.74\text{\AA}$  towards each other. This conclusion also has been reached by Richards<sup>13</sup> in molecular dynamics study using phenomenological potentials.

We have repeated these calculations for D<sub>2</sub> in Li and Al metals modelled respectively by 10 and 6 metal atoms occupying bulk sites. In both cases, we see a minimum in the energy  $\Delta E(R)$  at  $R \sim 0.7\text{\AA}$ . Furthermore, the energies at distances  $R \lesssim 0.2\text{\AA}$  between D-atoms in Li and Al are again nearly identical<sup>9</sup> with that in free space. Since Li and Al are both free electron-like metals with effective mass  $m^*/m \approx 1.0$  and Pd is a transition metal, we could safely conclude, in general, that at very short distances ( $R \lesssim 0.2\text{\AA}$ ) the electronic structure of the host metals do not influence the interaction between D-atoms significantly.

Thus far we have treated both H and D as identical

particles. We do not believe that the energetics would be qualitatively different if one were to relax the Born-Oppenheimer approximation, i. e. treat the effect of nuclear motion on the potential energy. However, the different mass of H and D would give rise to different vibrational frequencies and zero-point amplitudes. Using the potential energy along the (100) direction in  $\text{Pd}_6\text{D}_2$  cluster (see Fig. 4), we have calculated the zero-point energies, vibrational frequencies, and amplitudes for H and D. These are 0.14eV,  $0.69 \times 10^{14}/\text{s.}$ , and 0.17Å for H and 0.10eV,  $0.49 \times 10^{14}/\text{s.}$ , and 0.14Å for D. These can be compared with the corresponding values in free space, namely, 0.29eV,  $1.41 \times 10^{14}/\text{s.}$ , and 0.12Å for H and 0.21eV,  $1.0 \times 10^{14}/\text{s.}$ , and 0.10Å for D. Thus the vibrational characteristics of H and D in Pd are not very different from those in free space. To aid in our discussion of the possibility of fusion of  $\text{D}_2$  in Pd, it is useful to keep the following facts in mind. (1) Pd is an fcc lattice with a lattice constant of  $3.89\text{\AA}$ . (2) Deuterium in Pd would prefer to occupy the octahedral (O) sites until the stoichiometric limit of  $\text{PdH}_{1.0}$  is reached. The distance between two D-atoms occupying the nearest O-sites is then  $2.75\text{\AA}$ . (3) Under continued electrolytic charging, additional D-atoms could occupy the metastable tetrahedral (T) sites. The distance between D-atoms occupying the nearest T and O sites is  $1.68\text{\AA}$ . There may, however, be other metastable sites which can be occupied by the D-atoms.

Our results lead us to the conclusion that it is highly

unlikely that two D-atoms in Pd could be brought together to a distance of about  $0.3\text{\AA}$  to significantly enhance the fusion rate. In addition, the interaction between the two D-atoms in Pd at short distances is very similar to those in free space. The reasons for the excess heat reported by Pons and Fleischmann, therefore, have to be found elsewhere.

This work was supported in part by a grant from the Department of Energy (DE-FG05-87ER45316). One of the authors (P.J.) is thankful to Dr. F. Y. Fradin and Dr. K. S. Singwi for arousing interest in this calculation. We are grateful to Drs. B. Dunlap and C. White for pointing out the sensitiveness of the DVM code in determining energies and for sharing their results prior to publication.



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9. If anything, the D-atoms at distances  $R \lesssim 0.2A^{\circ}$  are actually more repulsive in a metal than they are in free space.
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12. B. I. Dunlap (private communication)
13. P. M. Richards (private communication).

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## Figure Captions

- Fig. 1. Cluster model for D-D interaction in Pd. The solid large circles are 6 Pd atoms taken to model the Pd bulk. The small solid circles are the two D-atoms equally spaced with respect to the octahedral site. The directions (100) and (111) along which the D-atoms were moved are also indicated.
- Fig. 2. Comparison between the energies of the  $D_2$  molecule (see Eq. (4)) as a function of interatomic separation obtained using Hartree-Fock (solid line) and local density approximation (solid triangles).
- Fig. 3. Interaction energy,  $\Delta E$  (see Eq. (5)) as a function of interatomic distance between D-atoms moving along the (100) (solid squares) and (111) (solid triangles) directions in Pd. The solid line represents the  $D_2$  interaction in free-space.
- Fig. 4. The energies in Fig. 3 around the range  $R \sim 0.7A^0$  is shown in an amplified scale. The legend is the same as in Fig. 3.

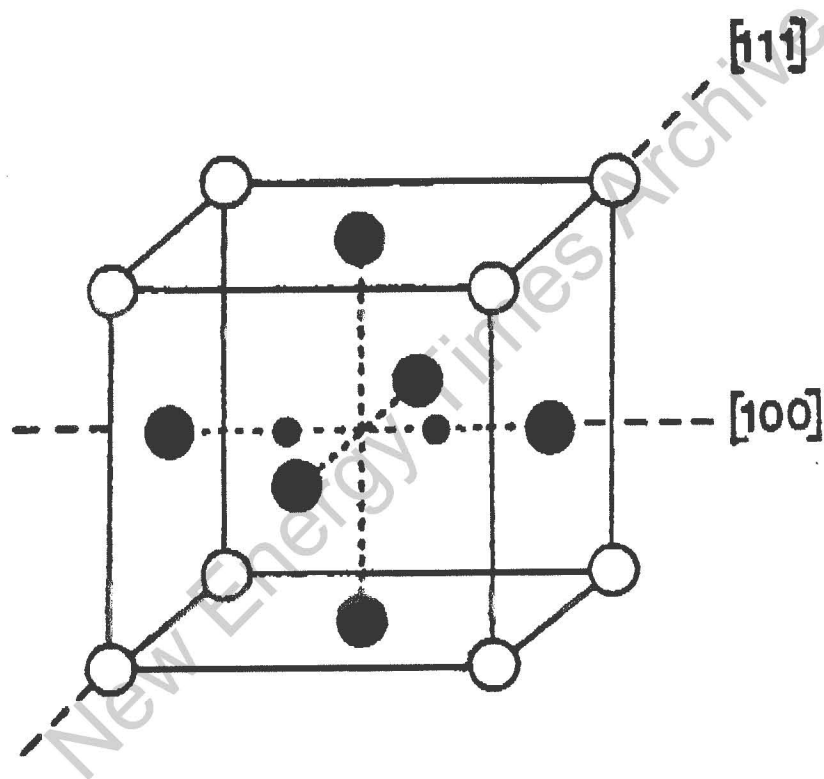


Fig. 1

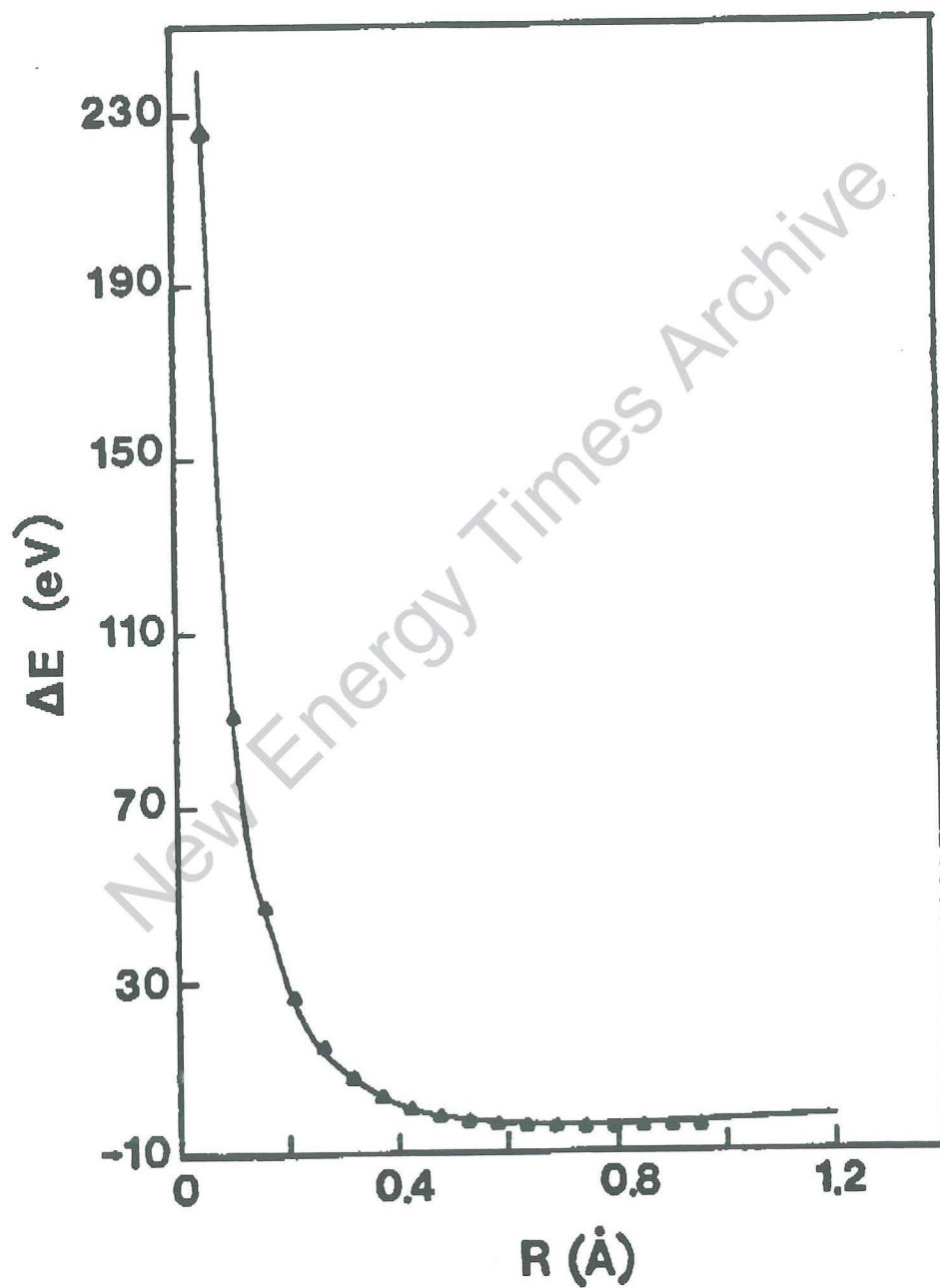


Fig. 2

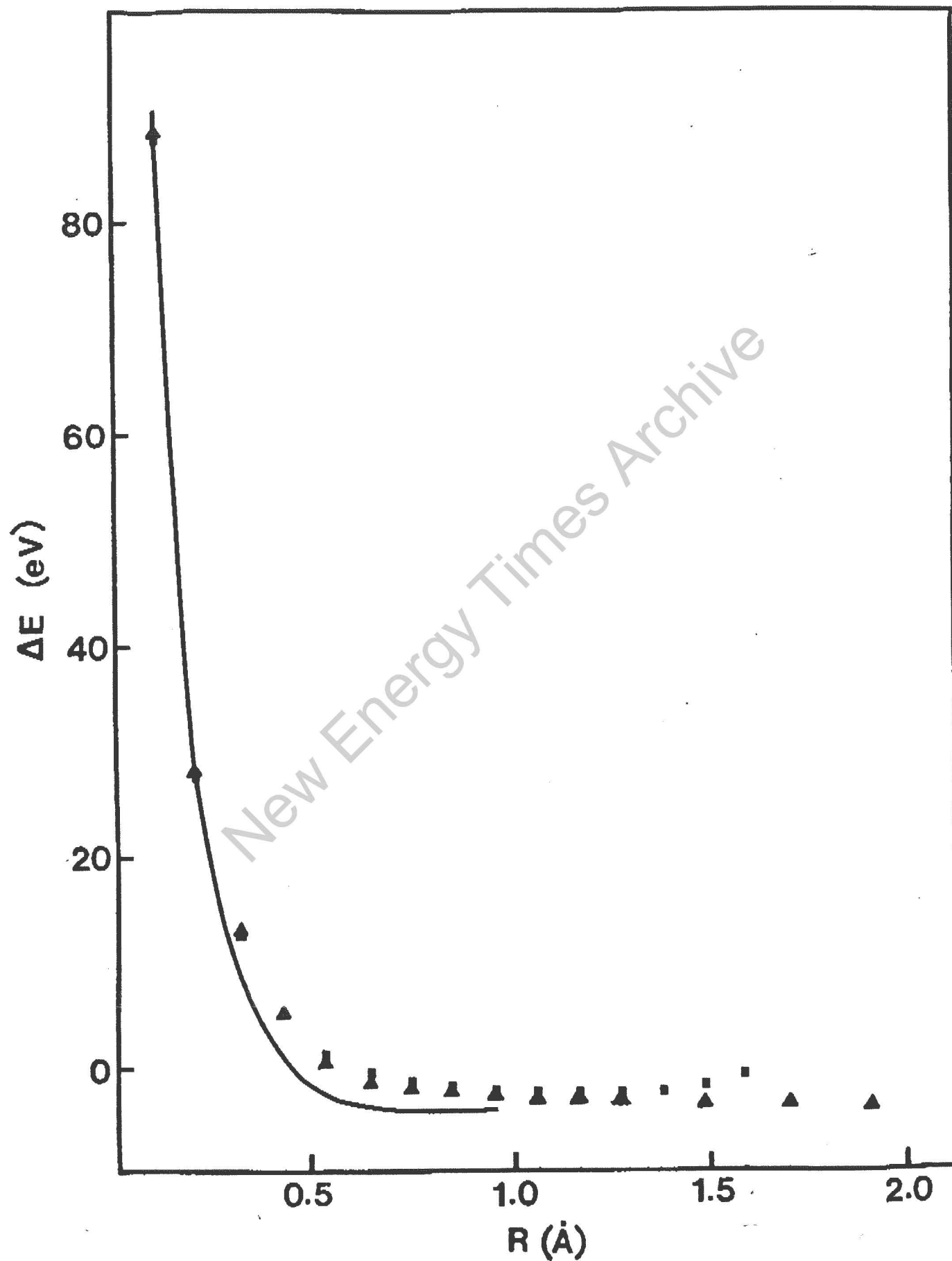


Fig 3

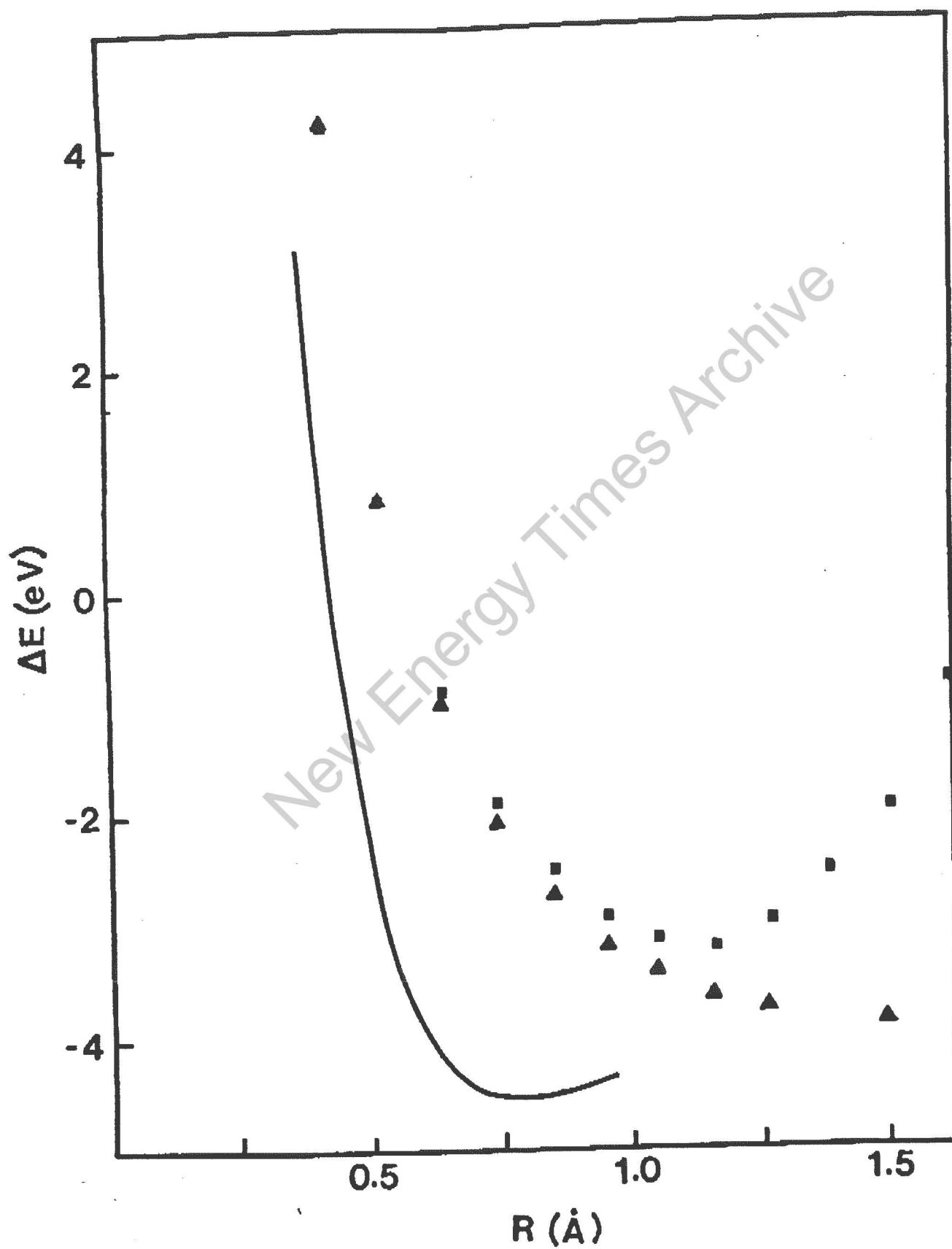


Fig. 4

To: Dr. William L. Woodard (Via FAX to 9 (202) 586-3119)

Richard L. Garwin  
IBM Research Division  
Thomas J. Watson Research Center  
P.O. Box 218  
Yorktown Heights, NY 10598  
(914) 945-2555

September 12, 1989  
(Via FAX to 9 (202) 586-3119)

Dr. William L. Woodard  
Cold Fusion Panel Secretary  
Energy Research Advisory Board  
to the  
United States Department of Energy  
1000 Independence Avenue, S.W.  
Washington, DC 20585

Dear Bill,

In preparing to write my section on neutrons in the "fusion products" part of the report, I have just spoken with Dr. Marcello Martone of Frascati. You remember that I visited Dr. Martone in his laboratory 06/09/89, to follow-up on the beautiful results of April on neutron bursts, and on the continuous neutron emission over 16 hours that were presented in the Frascati preprint by Scaramuzzi and co-workers. This work was also presented at the Santa Fe conference.

Although Dr. Martone returned from his vacation only yesterday, and so may not be up on the very latest results, he tells me that they have had no neutrons from their "dry fusion" experiments since those first results in April.

Martone says also that they shut off their electrochemical cells in July, having had no positive results from them either.

Martone had not heard of the work at BARC. I promised to tell him what I learned from my Telex communication with Dr. Iyengar at BARC, and Martone promised to send me by FAX any positive results from dry fusion from Frascati.

I have just talked with J.E. Schirber, lead author on the 06/28/89 paper "Search for Cold Fusion in High Pressure D2 Loaded Ti and Pd Metal and Hydride," submitted to Fusion Technology. In this and several papers, the group at Sandia National Laboratories found no positive results. Their background is much lower than that of Howard Menlove, and this discrepancy between Sandia and LANL was the origin of my communications with Menlove, resulting in the telephone conversation reported in my letter to you of 07/24/89. Schirber says they have only negative results at Sandia. In addition to the dry fusion work, they have consulted closely



with Steve Jones and tried all kinds of electrolytes. Schirber emphasizes that in one of their papers they report a run in which there was "an event for four hours" which was just like events reported as positive evidence by other groups. However, because Sandia had three channels of neutron counters, and two of them saw nothing, the one channel that did report counts must have been in error.

I have heard informal comments that several of the people who work with Howard Menlove do not believe that they are counting neutrons.

Schirber did not know of the BARC work. I am sending him a copy of the preprint:

Dr. Joseph E. Schirber  
Sandia National Laboratories  
Albuquerque, MN 87185  
W: (505) 844-8134  
FAX: (505) 846-2009

For further reference, the telephone number for Dr. Michael A. Butler is (505) 844-6897 and that for Dr. Ronald I. Ewing is (505) 846-4353. According to Schirber, Ewing (like Menlove) is an expert on neutron counting for treaty verification.

Sincerely yours,

Richard L. Garwin

RLG:jah:255&WLW:091289.WLW

**Energy Research Advisory Board**  
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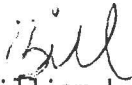
September 14, 1989

Subject: Chairman's July 20 Request for Summaries of Past and Present Cold Fusion Research and August 9 Request for Tritium Production Results

The following have been received to date from the following individuals in response to the above requests:

- R. Texas A&M University (response to August 9 request)
- S. T.F. Droege (response to July 20 request)
- T. S. Lien, Idaho Operations Office (response to August 9 request)
- U. M. Wadsworth, U. Utah (response to August 9 request)
- V. J. Farny, Institute of Plasma Physics, Warsaw (response to July 20 request)

Immediately behind this memorandum is a cumulative index.

  
William L. Woodard  
Panel Secretary

Enclosures

## INDEX

- A. Bo U.R. Sundqvist, Uppsala University (response to both requests)
- B. W. Scheid, Giessen University (responses to both requests)
- C. M.M. Broer, AT&T Bell Laboratories (response to August 9 request)
- D. J. Eridon, Naval Research Laboratory (response to August 9 request)
- E. Y.E. Kim, Purdue University (response to July 20 request)
- F. A Schriesheim, Argonne National Laboratory (response to July 20 request)
- G. M.A. Prelas, University of Missouri, Columbia
- H. L.R. Greenwood, ANL (response to August 9 request)
- I. F. Besenbacher, University of Aarhus (response to July 20 request)
- J. N.J.C. Packham (for Bockris), Texas A&M (response to August 9 request)
- K. J. Paquette, Atomic Energy of Canada, Ltd.
- L. N. Lewis, California Institute of Technology (response to August 9 request)
- M. J. Rafelski, University of Arizona (response to August 9 request)
- N. P.D. Bond, Brookhaven National Laboratory (response to August 9 request)
- O. J.L. Straalsund, Battelle (response to August 9 request)
- P. A. Narath, Sandia National Laboratories (response to August 9 request)
- Q. W. Appleton, Oak Ridge National Laboratory (response to August 9 request)
- R. Texas A&M University (response to August 9 request)
- S. T.F. Droege (response to July 20 request)
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- V. J. Farny, Institute of Plasma Physics, Warsaw (response to July 20 request)

## TEXAS A&amp;M UNIVERSITY

DEPARTMENT OF CHEMISTRY  
COLLEGE STATION, TEXAS 77843-3255

September 11, 1989

(409) 845-2011  
FAX (409) 845-4719Energy Research Advisory Board  
US Department of Energy  
1000 Independence Avenue, S.W.  
Washington, D.C. 20585

Please Note:

This short report describes our work involving the monitoring of tritium levels in heavy water electrolytic cells. The results are provided in numbered sequence as per the letter from John R. Huizenga of 8-9-89.

*C.R. Martin*1. Source and specific activity of D<sub>2</sub>O-

We obtained our D<sub>2</sub>O from two sources. The first source was Isotec, Inc. This D<sub>2</sub>O is 99.9% pure and has background tritium counts of  $20.7 \pm 1.0$  dpm/mL. Isotec D<sub>2</sub>O was used in 85% of our electrolytic cells. Aldrich 99.9% D<sub>2</sub>O was used in several of our first electrolytic cells. The background tritium activity of Aldrich D<sub>2</sub>O was measured at  $121 \pm 5.1$  dpm/mL.

2. Description of electrolytic cells, including volume of D<sub>2</sub>O-

Our electrolytic cells have gone through several phases of construction. Initially, we built ten cells in jacketed beakers to allow water cooling during the application of high current densities. Water cooling was used to minimize etching and dissolution of glass observed in several attempts at measuring gamma-rays and neutrons in cells which stayed at or near 100°C for several days. The cell volumes were approximately 200 mL.

The next set of cells avoided glass altogether. We built eight cells in 50 mL polyethylene and polymethylpentene centrifuge tubes to avoid high silicon concentrations observed using ICP analysis of the electrolyte from glass cells.

However, noting the announcement of tritium production in glass cells by Professor Bockris at Texas A&M, we built twelve more cells in

glass containers. Larger volumes were used, attempting to maximize the time between  $D_2O$  additions required by electrolytic and evaporation losses. These cells were built in 50 mL centrifuge tubes (8) and 30 mL specimen tubes (4).

We then built four closed cells using fuel cell electrodes as recombination catalyst. We hoped to increase sensitivity by avoiding possible tritium losses in the evolved gases. The cell volumes were 25 mL.

Lastly, in order to reduce minor differences between our cells and others at Texas A&M reported to produce tritium, we built twenty cells that to the limits of our abilities exactly duplicate this design. Six of these cells were run in light water as blanks. The pyrex centrifuge tubes have a volume of 15 mL.

All of the above cells were designed with the Pd cathode in the center, surrounded by the anode material. The anode material was a Pt wire coil in the first ten cells, Ni wire in the next 10 cells, and Ni gauze obtained from Prof. Bockris in the remaining cells. The cells were connected to DC power supplies in series, typically with 4-6 cells per supply depending on voltage limitations. Most of our cells (over 45) were built using 1 mm 99.9% Pd cathodes obtained from Hoover and Strong, the same source as the wire reported to produce by Professor Bockris. Other cells used Pd cathodes from a variety of sources with dimensions varying from 1 to 6.5 mm. The cells were sealed with septa in most cases. A sterile syringe was inserted allowing escape of evolved gases while minimizing exposure to  $H_2O$  in the air.

### 3. Cell Operating Conditions-

In all cases we operated the cells at low current densities, typically 60 mA/cm<sup>2</sup>, for an initial charging period averaging three weeks. Current densities were calculated assuming the geometric area only (no roughness factor accounted for). After this time, the cells were taken to higher current densities, typically 600-1000 mA/cm<sup>2</sup>. The water cooled cells were sustained at these current densities for several weeks while smaller cells were given higher currents for periods of 8-10 hours.

### 4. Schedule of $D_2O$ additions-

The schedule of  $D_2O$  additions depended on the volume of the cell. In the case of the 250 mL water cooled cells,  $D_2O$  was added (10-20 mLs) once a week during charging, and every three days during high current applications. Fifty mL cells required  $D_2O$  additions (3-5 mLs) every 3-4 days during charging and twice every eight hours during high current

applications. Smaller 15 mL cells required addition of D<sub>2</sub>O (1-2 mLs) every other day during charging and four times every eight hours during high current applications.

#### 5. Tritium Analyses-

Samples were withdrawn from working cells using a new sterile syringe for each assay. No purification procedure was used. LiOD concentrations in our cells were typically 0.1 to 0.2 M, resulting in little or no problem with chemiluminescence (CLM). Any sample showing CLM greater than 10% (less than one in 30 samples) was rejected. The method of analysis was Liquid Scintillation Counting (LSC) using a Wallac LKB 1219 for the first eight weeks and a Wallac LKB 1410 for the last three months. A water soluble scintillation cocktail (Biosafe II, Research Products International) was added to 0.1 to 1.0 mL of sample. LSC was performed in 10 steps of counting for two minutes each. The mean activity  $\pm$  one standard deviation was determined from this data accounting for volume and instrument efficiency.

Cells were assayed weekly during charging, and every two hours during high current applications. All the cells we have built show activity at this time between 25-45 dpm/mL. We attribute the slight increase in activity above Isotec D<sub>2</sub>O (background = 20.7 dpm/mL) to the natural <sup>2</sup>D-<sup>3</sup>T separation factor observed at Pd.

#### 6. Neutron production rate-

Professor Kevin Wolf has measured the neutron production rate in a number (about 15) of separate cells we built and operated at the Cyclotron Institute at Texas A&M. We have seen no evidence of neutron or tritium production from any of these cells.

#### 7. Heat balance-

Tritium levels were measured in fifteen operating calorimetric cells. No evidence of tritium production was found, even in two cells where excess heat was observed.

#### 8. Tritium sources-

To our knowledge, there are no tritium sources in or near our laboratory.



Thomas F. Droege  
2 S. 942 Thornecrest Lane  
Batavia, IL 60510  
6 September, 1989

John R. Huizenga  
Co-Chairman  
Cold Fusion Panel  
Energy Research Advisory Board  
United States Department of Energy  
1000 Independence Avenue, S. W.  
Washington, D.C. 20585

Dear Mr. Huizenga

Thank you for allowing me to comment on anomalous heat a.k.a.  
"Cold Fusion".

The following is in response to your request for a short summary  
of our work:

Researchers: Lee John Droege, Metallurgical Engineer  
Thomas F. Droege, Electrical Engineer  
About 10 other advisors and part time helpers

Sponsor: Gentlemen Scientists All, financed in part by  
wicked speculation in "Three Mile Island Stock"  
bought at 4 1/2, sold at 32. We had faith in the  
need for nuclear power.

Apparatus: Closed Integrating Electronic Calorimeter of our  
design. Catalyst in cell completely recombines  
electrolysis products. Any unbalance, as caused by  
absorption of deuterium into the palladium, is  
measured in a gas burette. A four terminal  
impedance connection monitors the phase change of  
the palladium while in the cell. Safety is  
monitored by a 1" NaI(Tl) Scintillation Counter  
connected to the data acquisition system. A twelve  
bit high speed data acquisition system monitors 17  
parameters, including self test features. System  
records the average of 500 samples of each variable  
each minute.

Calibration: Calorimeter stability has been measured to +/- 1  
milliwatt. The calorimeter has a capacity of 7  
watts. We have not yet had time to achieve  
ultimate calibration since a suitable experiment  
will take about one month. Present calibration  
good to +/- 15 mw.

Results: Approximately the right amount of (presumed) oxygen  
appears in our burette to match the D which has



gone into the palladium. This is confirmed by impedance change. Excess heat appears when gas is absorbed to near the .7 D/Pd ratio. We have runs where excess heat (we measure Joules) has accumulated to the extent of 20 ev per palladium atom. A typical run accumulated 10,000 J of excess heat over 50 hours. The detailed structure of the high speed measurements is very convincing that something other than calorimeter drift is occurring. We are about six months of careful calibration away from publication.

Comments: We think that any conclusion in this field is premature. These experiments are very difficult. Our calorimeter takes about 10 hours to settle to 0.1%. The slightest mistake causes the loss of many time constants (read days) work. We have made many mistakes.

The following is not for publication:

We wish we were perfect like Dr. Lewis and could come to a conclusion after a few days work. We fear that we will have to grub on for many months before we can state our results with certainty. We started building a calorimeter as soon as we saw the McNiel - Lehrer report. We put the first Pt-Pd-D sample in it on July 22. Every run since has been less than perfect. Something always goes wrong, like we tried nickel wire for the anode to save money, and the cell turned green (but we got a good measurement on what a chemical reaction looks like).

Now I believe that I have a certain objectivity in that I am not asking for money. I have sympathy for Pons and Fleischmann since I believe their cover was blown prematurely. Now that I have been trying to do the experiment I can easily believe that it took them five years to get to where they are. People who express horror that Pons and Fleischmann didn't do such and such just don't know what they are talking about. I have a list that could take years to work through of things that are obvious to try. I don't need Dr. Lewis to come around and ask sneeringly why I have not done Pt on Pt with D2O or some such other obvious experiment, and then look knowingly wink-wink at the crowd.

So what you are doing is driving the work elsewhere. This means that in the end, you will not know what is going on. If it proves to be an important discovery, then the DOE will look like fools, and will not have any of the stars in their stable. Believe me, some are taking a chance and putting the good workers under contract.

Many years ago when I worked at the Princeton-Penn Accelerator I occasionally had contact with the Princeton Plasma Fusion group with which we shared the site. They were turkeys then, and I

presume they have not changed. So it looks a lot to me like they are trying to protect their turf by putting down the "anomalous heat" work. OK, they have had their billions and 20 years to do something. I see zero chance that any of their schemes will work. Why not encourage new ideas? Some places like Stanford and Texas A&M look like they are doing good work. Why not support a few students? I am doing just fine on my own, though I am offended by having to pay \$1.50 per inch for #30 platinum wire.

I recently participated in a summer study at Breckenridge CO where over 350 high energy physicists from all over the world participated. I made it a point to talk to as many as I could about "anomalous heat". You would think I was ringing a bell and crying "leper, leper". Still I managed to corner about 40 and made them talk to me. About 35 had completely closed minds, and quoted to me second and third hand hearsay that they claimed proved that "cold fusion" was wrong. Since I had copies of the papers they were claiming to quote it was easy to detect their errors. When called on their errors they always quoted someone else that they trusted. I found very few nay sayers that had read the papers.

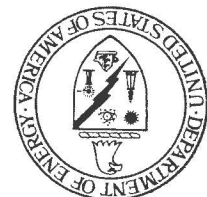
So the conclusion of your committee is not surprising. Collect 22 "scientists" off the street and 20 of them will have minds closed to new ideas. The 20 quickly beat up the two and a report is produced like you sent.

Still I found a few open minds, even a famous theorist who was ready to think about a new field (other than e&m). Page 3 of your report states "Since deuterium fusion necessarily yields fusion products ...". Not so necessary my friends say (when only slightly drunk). Unless you extend gamma rays into the infra red. If you have read the Texas A&M preprints you get very excited when an old medium energy physicist friend says that  $6\text{Li} + \text{D} \rightarrow 2\alpha + \text{heat}$  is possible. The branching ratios often quoted are taken under very special circumstances. There is no reason to believe that they apply in the palladium lattice.

You are setting the DOE up for a neutral-lose game when you could have a neutral-win game by keeping an open mind and delaying the taking of a position until there is sufficient time for those who are doing careful experiments to publish. While Admiral Watkins may want a quick answer, it is going to take years to sort this out. It may be another poly water, but I am betting the rest of my productive life that it is not.

very truly yours,

  
Thomas F. Broege



Department of Energy  
Idaho Operations Office  
785 DOE Place  
Idaho Falls, Idaho 83402

September 5, 1989

Dr. William Woodard  
ER-6, 3F-061  
Office of Energy Research  
1000 Independence Avenue, SW  
Washington, DC 20077-9381

SUBJECT: "Cold Fusion" Results Summary

Dear Dr. Woodard:

In response to the request in the letter from Dr. John R. Huizenga to INEL Manager, Don Ofte (dated August 9, 1989), I am including the following two documents (Enclosures 1 and 2) for your information.

Enclosure 1 is a summary of cold fusion results from INEL. It provides brief answers to the specific questions contained in the aforementioned letter from Dr. Huizenga, including the result of tritium measurement during an electrolysis of D<sub>2</sub>O. Enclosure 2 is a preprint of a presentation by G. R. Longhurst from INEL at the Workshop on Cold Fusion Phenomena held at Santa Fe, New Mexico on May 23-25, 1989. I believe these two documents should describe INEL's limited activities in the cold fusion phenomena. If more information is needed, or if you have questions, please do not hesitate to contact me at (208)526-1231 or Dr. (Gus) A. J. Catfrey at (208)526-4024.

Sincerely,

Stephen C.T. Lien, Chief  
Advanced Technologies Branch

Enclosures

cc: Dr. Jacob Bigeleisen

Department of Chemistry

SUNY at Stony Brook

Stony Brook, NY 11794-3400

# COLD FUSION RESULTS SUMMARY

G. R. Longhurst and A. J. Caffrey  
Idaho National Engineering Laboratory  
EG&G Idaho, Inc.  
P. O. Box 1625  
Idaho Falls, ID 83415  
August 28, 1989

Integrated "cold fusion" experiments were conducted at the Idaho National Engineering Laboratory (INEL) during the period April 4 to May 18, 1989. The objective was to attempt observation of radiation emission and/or thermal energy. No positive results were observed. A description of the work is available in Ref. 1. The following summary facts are provided in accordance with the request in Ref. 2:

The electrolytic cell is shown schematically in Figure 1. The cell was housed in a 50-ml Pyrex test tube. Electrolyte volumes were 30 or 40 ml. Waters of varying compositions were used.  $D_2O$  used was from a stock on hand at the INEL for approximately 20 years. Nominally considered "high purity", it had been stored in sealed glass flasks. No compositional analysis was done except to measure the tritium activity level on one 10-ml sample from the stock flask using liquid scintillation counting (Packard Model 2250-CA scintillation counter, 20-ml total sample volume - 50% Packard Picofluor scintillation cocktail in polyethylene vial). Tritium activity in that sample was  $1.59E-04 \pm 2.77E-06 \mu Ci/mL$ .

The cathode used in most experiments was a coil of 1-mm diameter Pd wire. The initial length of the wire was 14.5 cm. It was coiled to a helix nominally 1 cm in diameter and 4 cm long. Other cathodes used included a straight rod of the same Pd wire and a Pd foil 0.05 cm thick, 1 cm wide and 5 cm long. The wire was purchased from Morton Thiokol Alfa Products (Puratronic grade) and used without further processing except surface cleaning by washing in solvents. The foil was from commercial grade stock on hand. The anode was a 30-cm length of 0.5-mm diameter Pt wire coiled to fit against the walls of the test tube.

The cell was operated at a variety of currents, but most measurements were made at currents of 200 or 400 mA. The voltage required for operation



depended on the electrolytic composition, electrode geometry, and cell temperature. Basic operation was at room temperature, but heat measurements were made by encasing the test tube with a polystyrene foam block whose thermal conductance had been measured by replacement of the electrodes with an electrical resistance and tracking the temperature transient as well as by observing the temperature difference of the cell contents with the outside when it was in place.

LiOH salt was dissolved in the water. Most measurements were made with 0.1 Molar salt solution for electrolyte, but measurements at higher and lower concentrations were also made. Unmixed  $D_2O$  was used in some tests, purified  $H_2O$  in others, and a mixture of the two was also tried.

Only one attempt was made to observe tritium production. In a test that ran about 5.7 days,  $D_2O$  was added twice during the test to bring the electrolytic volume back to 40 ml for an average consumption rate (electrolysis of  $2.58 \pm 0.1$  ml/day plus evaporation) of  $3.0 \pm 0.2$  ml/day. The tritium activity of a 10-ml sample from that cell was measured using an identical procedure to that discussed previously and found to be  $1.67E-4 \pm 2.92E-06$   $\mu\text{Ci}/\text{ml}$  (a 5% increase). A cursory theoretical analysis suggested that the amount of increase in the tritium level of that sample relative to the original stock  $D_2O$  was, within experimental error, just what would be expected due to the concentrating effect of the electrolysis. The only tritium source operating in the vicinity of the experimental laboratory was the Advanced Test Reactor located approximately 500 m away in a direction perpendicular to that of the prevailing winds. It releases approximately 12 Ci/month in water, but there is no measurable tritium escape into the air nor background tritium concentration in the laboratory that would influence the present results.

Neutron and gamma ray measurements were made during these experiments. A large scintillation detector, 12.7 cm in diameter by 12.7 cm long, filled with Bicron BC-501 liquid scintillator was installed about 20 cm from the test cell. A Nuclear Data ND-66 multichannel analyzer (MCA) was used for analysis. The counting system gain was adjusted with a  $^{22}\text{Na}$  source to 1.75 Mev electron-equivalent ( $\text{MeV}_{ee}$ ) at full scale. At this gain the prominent

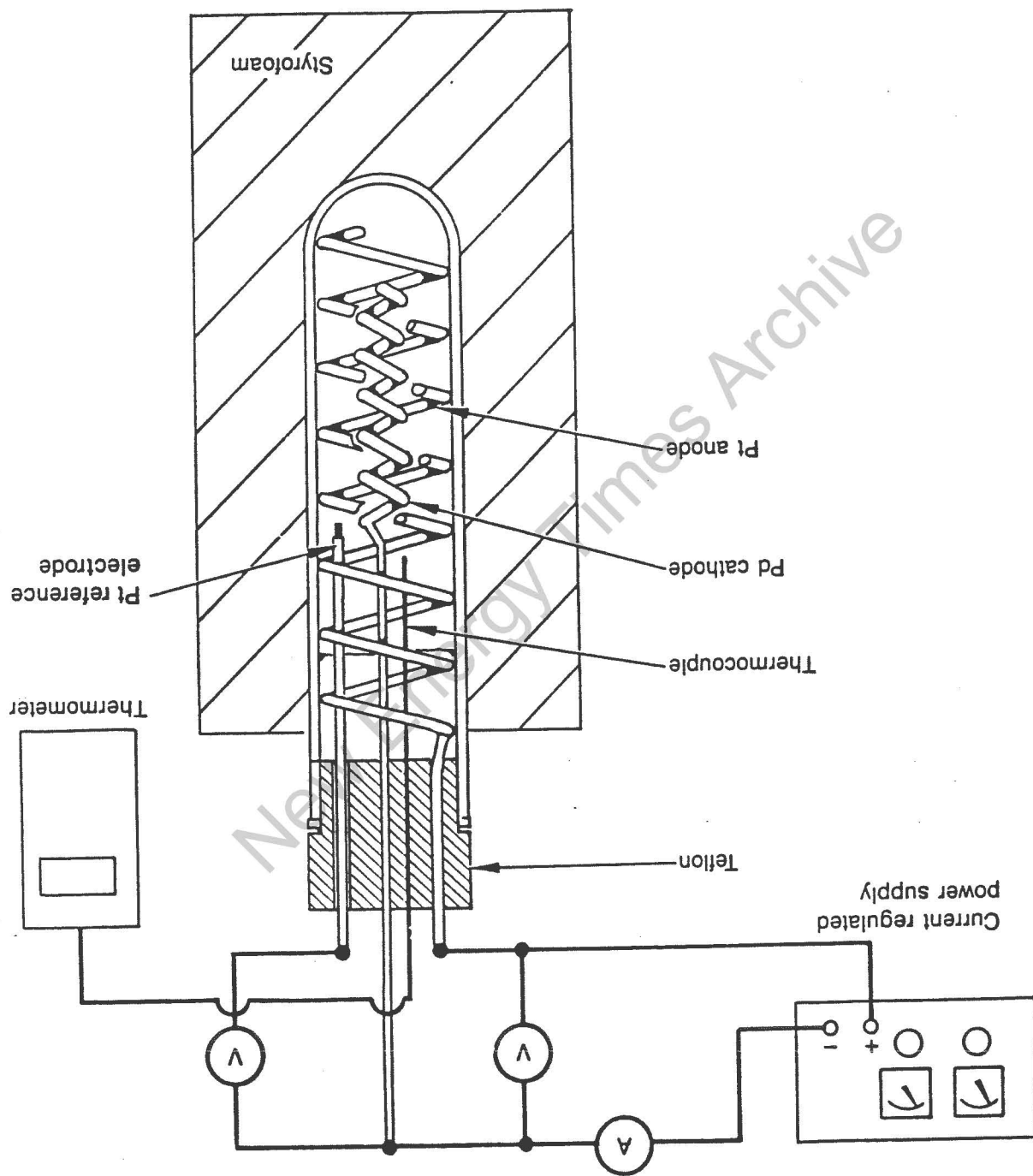
"edge" of the recoil proton distribution of 2.5 MeV fusion neutrons would have appeared in the middle of the pulse-height spectrum, at about 0.9 MeV<sub>ee</sub>. As a check on the detector sensitivity, a small ( $10^4$  n/s)  $^{252}\text{Cf}$  source was introduced at the same distance from the liquid-scintillation detector as the test cell. Count rates observed then were 30 - 60 times background rates, and there was a notable shift in the spectrum of energies indicated by the MCA. There were no other indications of radioactivity other than background during the entire course of the electrolysis experiments.

Heat balances were made by careful observation of temperature differences during periods when the cell was insulated by the foam block to a constant velocity and temperature air stream. Type K thermocouples were used with a sensitivity of 0.1 K. By comparing temperatures achieved during these measurements with those with a known power input, thermal fluxes through the cell could be measured to a precision of less than 40 mW. No net production or consumption of energy was observed within that limit while the cell was operating in steady-state.

## REFERENCES

1. G. R. Longhurst, T. J. Dolan, and G. L. Henriksen, "An Investigation of Energy Balances in Palladium Cathode Electrolysis Experiments," EGG-M-89203, May 19, 1989, EG&G Idaho, Inc.
2. J. R. Huizenga letter to D. Ofte, August 9, 1989.

Figure 1. Schematic of the experimental apparatus used.







**Idaho  
National  
Engineering  
Laboratory**

**EGG-M-89203  
PREPRINT**

**AN INVESTIGATION OF ENERGY BALANCES IN  
PALLADIUM CATHODE ELECTROLYSIS EXPERIMENTS**

**G. R. Longhurst, T. J. Dolan, G. L. Henriksen**

**May 19, 1989**

**Workshop on Cold Fusion Phenomena,  
May 23-25, 1989, Santa Fe, NM**



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# AN INVESTIGATION OF ENERGY BALANCES IN PALLADIUM CATHODE ELECTROLYSIS EXPERIMENTS\*

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Idaho National Engineering Laboratory  
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Idaho Falls, ID 83415-3523

## ABSTRACT

In recently publicized cold fusion experiments at the University of Utah, generation of excess heat was reported. To investigate mechanisms that may contribute to energy flows in electrolysis cells, a series of experiments was performed at the Idaho National Engineering Laboratory (INEL). Ordinary water ( $H_2O$ ), heavy water ( $D_2O$ ), and a mixture of the two were used in the INEL experiments. Cathodes used include a 51- $\mu m$  Pd foil and 1-mm diameter extruded wire Pd rods in two configurations. Energy balances in these experiments revealed that some of the required voltage to sustain a given current is due to irreversibilities associated with cell operation. Particularly significant are electrolyte resistance and activation energy polarization effects. Energy balances in the INEL experiments showed there was no significant net gain or net loss of energy. Cell overpotential curves were fit well with a Tafel equation, with parameters dependent on electrode configuration, electrolyte composition, and temperature. Water evaporation and interactions of hydrogen isotopes with the Pd cathode were evaluated and found not to be significant to energy balances. No ionizing radiation, tritium production, or other evidence of fusion reactions was seen in the INEL experiments.

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\* Work supported by U.S. Department of Energy, Director of Energy Research, under DOE Contract No. DE-AC07-76ID01570.

## INTRODUCTION

In recent experiments reported by Fleischmann and Pons<sup>1</sup> and widely discussed, excess heat production was reported that has stimulated great interest in the "cold fusion" process as a potential energy source. Energy production several times the input energy was reported leading to consideration of energy break even and gain. The published rates of energy release were inconsistent with the rates of conventional fusion reactions suggested by reported neutron and gamma ray detection.

To better understand the processes dominant in the operation of a cell of the type described by Fleischmann and Pons, a series of Pd-cathode electrolysis experiments was conducted at the INEL. The main technical objectives considered in these experiments were the evaluation of energy flows and the verification and measurement of any nuclear reactions.

## EXPERIMENTS

An apparatus shown schematically in Figure 1 uses a 40-mL Pyrex test tube, nominally 2.5 cm in diameter. One Pd cathode used in these experiments was a foil 51  $\mu\text{m}$  thick, 1 cm wide and 5 cm long. At one end of the foil a 0.2-mm diameter copper wire was laser-beam welded. Other cathodes used were made of 1-mm diameter drawn Pd wire. One of these was a straight rod extending about 8 cm into the electrolyte. Others were 14.5-cm lengths of Pd wire coiled into a 1-cm diameter helix approximately 4 cm long. The manufacturing process for the Pd (drawing vs casting) should not be particularly significant because the phase change that occurs on hydriding will mostly obliterate the preexisting grain structure. The coil cathodes were fully immersed in electrolyte suspended by welded Cu or Pt lead wires. The rod was simply inserted into the

electrolyte as far as it would go. The foil cathode was operated fully immersed in some tests and in others only partly immersed. The anode was a 0.5-mm diameter Pt wire 30 cm long that was coiled to fit against the wall of the test tube. On some tests an additional Pt/H<sub>2</sub> reference electrode was provided to study individual electrode potentials and quantify overpotentials associated with each electrode. This consisted of a 0.5-mm diameter Pt wire encased but for 1 mm at the tip by heat-shrinkable plastic. The tip of the reference electrode could be located at arbitrary positions in the cell.

Mechanical support, thermal insulation, and vapor exchange suppression were provided by a plug of styrofoam (nominally 10 cm long) or Teflon (2.5 cm long). A 0.5-mm (0.020-inch) diameter, Inconel-clad, type K (chromel-alumel) thermocouple was inserted through the plug. Additional thermal insulation was provided by a styrofoam block, nominally 5 cm x 10 cm x 15 cm shaped to accept the test tube. A second thermocouple was located in the air about 5 cm from the assembly. Various instruments were used for measuring currents and voltages. Most tests were performed using Fluke 8050A digital multimeters. Temperature measurements were made with Fluke 52 K/J thermometers. Since only differential temperature measurements were important, it was sufficient to ensure that these agreed with each other within 0.1 K. Power supplies included HP 6126 A and HP 6260 B units operating in the current-limited mode. The latter was used with a 0.5-ohm shunt resistor to improve regulation.

Thirty or forty mL of 0.1 molar (up to 1.2 molar in some tests) solution of LiOH in H<sub>2</sub>O or D<sub>2</sub>O or a mix of the two was placed in the test tube, and the electrode/thermocouple assembly was inserted. Sealing was not necessary because the tests were conducted in air. Currents up to 5.7 A were directed through the cell, with most tests run at 200-500 mA. Several types of tests were conducted: (1) calorimetry tests in which the temperature and applied voltage histories of the cell were monitored with constant current through the cell; (2) electronic resistor heating tests to measure the thermal conductance for each cell configuration used; (3) applied current vs cell voltage tests (generating cell polarization



curves) to measure equivalent resistance of the cell; (4) tests in which the cell was run at constant current, but the cell surroundings were changed to include a heater tape, the foam block, and a container of cold tap water to measure the temperature sensitivity of the cell potential characteristic; and (5) constant-current voltage buildup tests and open circuit potential decay tests to observe electrochemical transient phenomena. In some tests the reference electrode was moved to various locations inside the cell to observe potential variations in the electrolyte. In others, the interior thermocouple was swept over the length of the test cell to observe temperature variations.

## CONCEPTUAL APPROACH

### THERMODYNAMICS

Take as a control volume (C.V.) the volume bounded by the test tube and plug of Figure 1. Following standard sign conventions, consider  $\dot{W}$ , the rate at which work is done by the C.V.;  $\dot{Q}$ , the rate of heat transfer to the C.V.; and the flow term,  $\sum \dot{n}_i h_i$ , or the rate at which energy leaves the control volume via molar fluxes,  $n_i$ , and their associated partial molar enthalpies,  $h_i$ . Water in the C.V. is electrolyzed at the molar rate  $n_w$  with enthalpy of formation,  $-\Delta H_f$  ( $\Delta H_f$  values for water formation, the reverse of electrolysis, are tabulated in handbooks). The basic thermodynamic requirement is that

$$\dot{Q} - \dot{W} - \sum \dot{n}_i h_i = \dot{H}(\text{C.V.}) \quad [1]$$

Each of the terms in this balance is considered now in more detail.

#### Heat

Heat rate measurement is usually accomplished by observing a temperature transient in a system of known thermal capacity or looking for

a steady-state temperature difference across a known conductance. In the present work both methods were used. A small C.V. thermal capacity and a constant temperature heat sink were sought to eliminate the effects of time lags. A low thermal conductance enhances the sensitivity of the experiment for measurement of low heat rates. A nearly constant temperature reservoir was available in the air flow of the fume hood used for hydrogen safety. This had the additional advantage of not requiring sealing for water mixing. Ambient air temperatures usually varied less than 1 K over the course of a test, and the flow conditions were constant. A styrofoam insulation block was installed over the test cell during transients to reduce thermal conductance. In measuring that conductance it was straightforward to replace the electrodes in the test cell with a simple 5-ohm electronic resistor. By making measurements of the difference in temperature between cell contents and the surroundings as a function of time it was possible to determine the thermal conductance,  $\kappa$ , from the ultimate temperature difference,  $\Delta T_f$ , reached in steady state,

$$\kappa = \frac{\dot{W}}{\Delta T_f}, \quad [2]$$

where for steady-state temperature,  $T_f$  and ambient temperature,  $T_\infty$

$$\Delta T_f = T_f - T_\infty. \quad [3]$$

Using  $\kappa$  and the time constant,  $\tau$ , for approaching steady state, the thermal capacity,  $C$ , of the apparatus could be determined. Again for a fixed input power,  $\dot{W}$ ,

$$C = \tau \kappa. \quad [4]$$

Stirring is of considerable importance in experiments of this type. It was determined by measurement that when electrolysis was ongoing in the test cells, there was no detectable temperature difference (0.1 K resolution) over the cell volume except for a small region at the very bottom of the test tube. This mixing was facilitated by the stirring action of the bubbles which occupied the bulk of the cell length and the entire diameter, moving up near the electrodes and down in the annulus

in-between. It was further aided by thermally driven convection of the fluids in the cell, by the insulation provided by the styrofoam (causing essentially all of the temperature drop to occur outside the C.V.), and by the small volume and surface-to-volume ratio of fluid used.

In electronic resistor calibration tests there are differences in fluid circulation patterns from those during electrolysis. The cell is well mixed by the bubbles during electrolysis, but only thermal convective currents were active mixers during the calibration runs. However, temperature variations measured during electronic resistance calibration tests were not greatly different from electrolysis runs. Temperatures in the calibration tests were lower at the bottom of the cell than in the bulk, but over most of the cell volume they were constant to within 0.3 K.

The thermal conductance,  $\kappa$ , was found in these calibration tests to be quite constant with applied power to the resistor up to operating voltages where electrolysis took place on the bare leads. If undetected this would be a source of considerable error. This is illustrated by Figure 2 which shows the values obtained for the D<sub>2</sub>O cell with 30 mL of solution as a function of the potential applied to the resistor. The values above 2.2 V follow a different track because some of the power is being applied to the reversible electrolysis work for those data points. After this was noticed, subsequent measurements were made with insulated leads. Tests at zero potential were conducted by heating the entire cell, allowing it to cool normally in the air stream of the fume hood, and comparing time constants with those for powered, steady-state temperature difference measurements. Their slightly higher values show the effect of the cooler region at the bottom of the cell for the electronic resistor heated tests. Conductance values of 0.037 W/K were found for the 30-mL D<sub>2</sub>O experiments and 0.059 W/K for 40-mL D<sub>2</sub>O experiments. Similar measurements gave a value of 0.042 W/K for 30-mL H<sub>2</sub>O experiments. Those values were used in determining heat fluxes with the formula

$$\dot{Q} = \kappa \Delta T_f \quad [5]$$

As a check, on electronic resistor-heated calibration tests the thermal capacity, C, was determined from Eq.[3] and compared with values



calculated from tables of thermophysical data and measured masses of the cell components. The agreement was satisfactory (less than 3% error).

In each case the heat measurement was made at a temperature elevated 12 - 60 K from the ambient temperature. The temperature swing was desirable so that a transient could be observed. Note that the transient time constants in these tests were different from those in the electronic resistance calibration tests because of the sensitivity of applied voltage to temperature. From the time constant measured on these tests and thermal capacity measured previously, an additional estimate of thermal conductance was obtained from

$$\kappa = \frac{C}{\tau} + I\alpha, \quad [6]$$

where  $\alpha$  is the rate at which the required voltage changes with temperature according to

$$V = V(T_{\text{ref}}) + \alpha(T - T_{\text{ref}}). \quad [7]$$

These estimates agreed well with those made in electronic resistor heating tests.

### Work

The work crossing the boundary of the C.V. is mainly electrical, and the net work is negative. Flow work is done by the escaping gases, but it is accounted for by using enthalpy rather than internal energy in the flow term of Eq.[1]. The electrical potential,  $V$ , applied at the cell electrode junctions to sustain current,  $I$ , depends on a number of factors in addition to the electrolysis work, notably activation polarizations and resistive effects. These processes are dissipative, temperature sensitive, and geometry and electrolyte concentration dependent. These losses were found to be not negligible in energy balances for these experiments.

The potential difference between the electrodes is the sum of the reversible electrolysis potential (1.26 V for D<sub>2</sub>O), the resistive drop, and the activation overpotential, given by the Tafel equation,<sup>2</sup> which may be written in the form

$$V = [a + b \ln(I) + IR]_{\text{cath}} + [a + b \ln(I) + IR]_{\text{anode}} \quad [8]$$

The constants in Eq.[8] will depend on the specific configuration of the experiment.

### Energy Flux

Various processes connected with energy flows inside the cell and across the cell boundary were considered to evaluate their significance to an overall energy balance. These include (1) evaporation of water from the surface of the electrolyte and loss with the evolved gases, (2) reformation of water on the surfaces of the electrode leads above the electrolyte, (3) diffusion of hydrogen or deuterium into or out from the cathode during temperature ramps, (4) diffusion of hydrogen or deuterium in the cathode up through the surface of the electrolyte followed by its unrestrained release, and (5) Peltier and Thomson heat effects. These were each evaluated for the contribution they would make to the observed heat rates and found to be less than the level observable in the present experiments. Detailed discussion is presented in the Appendix.

If the gases exiting the cell during its operation are at the same temperature as the water from which they came, and if that is close to standard conditions of 1 bar pressure and 298 K, then their collective enthalpy is changed only by  $-\Delta H_f^0$  from what it was in the water. The enthalpy of elemental gases at standard conditions is usually defined to be zero. Hence, the flow term in Eq.[1] may be neglected provided that the enthalpy change is accounted for on the right side of Eq.[1].

## RESULTS

### Energy Balance

Table 1 lists a summary of experimental conditions and results of these experiments. Except as noted, the cathodes were fully immersed in electrolyte. Only magnitudes are listed in Table 1 for applied potentials and heat transferred. The heat increment listed is calculated from

$$\Delta \dot{Q} = \dot{Q} - I(V - V_{el}) \quad [9]$$

which is just a reformulation of Eq.[1] where  $V_{el}$  is the enthalpy change per electron incorporated in the electrolysis (1.53 V for  $D_2O$ , 1.48 V for  $H_2O$ ).  $\Delta \dot{Q}$  represents the imbalance in the heat observed during the experiment. The  $\Delta \dot{Q}$ s presented in Table 1 correspond to a 4.4% standard deviation in the heat fluxes measured with no particular sensitivity to water type, run time, electrode configuration or operating current. Slight changes in conductance from one test to another as suggested by the scatter in Figure 2 and the limit of 0.1 K in temperature measurement sensitivity could easily account for the incremental heat values observed.

### Irreversibilities

Figure 3 shows a typical voltage characteristic (required driving potential for a given current) for each electrode referred to the floating reference electrode potential. Note that a minimum voltage is required before any current flows (before any electrolysis takes place). This constitutes a back emf or reversible potential for the electrolysis of water. The theoretical value for this potential is 1.26 V for electrolysis of  $D_2O$  or 1.23 V for  $H_2O$ . As shown in Figure 3, the measured potential difference between electrodes at the lowest current values was close to that.

As current begins to flow, activation polarization potentials set in. These are due to irreversibilities associated with electron transfer

steps at the electrode surfaces. The stronger effect is at the anode and is due to complex processes associated with oxygen molecule formation. These processes have been studied extensively (c.f. Ref. 3). The smooth curves in Figure 3 are computed using Eq.[8] with the following constants:

	<u>Anode</u>	<u>Cathode</u>
a (V)	2.1	0.6
b (V)	0.04	0.07
R ( $\Omega$ )	3.4	3.0

The straight lines indicate the IR terms. Measurements of the spatial distribution of the potential by the floating reference electrode (Figure 4) and other measurements using a 1-kHz, ac impedance bridge connected to the electrodes confirm that the resistance is mainly in the electrolyte and that it is about 6  $\Omega$  for the cell configuration of Figure 1 when using D<sub>2</sub>O.

Table 2 shows a comparison of the combined constants of Eq.[8] to give the overall cell potential, obtained by fitting experimental data from several different experiments. Changes in electrolyte concentration only seem to affect the resistance, while changes in cathode configuration also influence the other coefficients, a and b.

Figure 5 shows the voltage onset characteristic for a coil cathode operated in D<sub>2</sub>O (a) with a similar curve for a foil cathode (b). In each case there is a cell operating voltage increase of approximately 0.5 V after start-up. In the foil tests, the time required to establish the voltage is a few minutes. For the coil-cathode tests it takes much longer, and it appears to come in two distinct steps. The first part of the transient is thought to be due to electrochemical processes at the electrode surfaces, principally at the anode. That should appear in about the same time for any cathode configuration. The second part is believed to be associated with x, the H/D-to-Pd atom ratio in the cathode. In the thin foil that appears to take but a few minutes. For the 1-mm wire, in either the rod or coil cathode configurations, the second step requires on the order of a day. The times required for the second step correlate well



with estimated diffusion times for hydrogen in the Pd ( $D = 10^{-7}$  cm<sup>2</sup>/s). The sudden change in potential at about 390 min Figure 5(a) represents a change in time scale and is to show the value achieved by the next day. This long-time-constant change was in the cathode. The anode voltage did not change in that time.

Using the apparent hydriding potential change of 0.3 V (the observed change in cathode potential over a 24-hour period) with the Fermi energy shift function for highly loaded ( $x > 0.6$ ) Pd given by Wicke and Brodowski,<sup>4</sup>

$$\Delta E_f(x) = 0.867 (x - 0.52) \text{ (eV/atom ratio)}, \quad [10]$$

it appears that loading up to  $x \approx 0.9$  may be occurring. It is noted from the logarithmic term in Eq.[A-4] in the Appendix that an infinite constraining potential would be required to achieve  $x = 1$ , assuming that equation is valid in that regime.

A typical open-circuit voltage characteristic is shown in Figure 6. The activity indicated by the sustained open-circuit voltage is mainly associated with complex chemical relaxation processes at the anode such as the formation of deuterium peroxide (half cell potential of 1.77 V) as the anode changes from an oxygen electrode to a hydrogen electrode.

### Detecting Fusion

**Direct Radiation.** Neutron and gamma ray measurements were made during these experiments. A large scintillation detector, 12.7 cm in diameter by 12.7 cm long, filled with Bicron BC-501 liquid scintillator was installed about 20 cm from the test cell. A Nuclear Data ND-66 multichannel analyzer (MCA) was used for analysis. The counting system gain was adjusted with a <sup>22</sup>Na source to 1.75 MeV electron-equivalent (MeV<sub>ee</sub>) at full scale. At this gain the prominent "edge" of the recoil proton distribution of 2.5 MeV fusion neutrons would have appeared in the middle of the pulse-height spectrum, at about 0.9 MeV<sub>ee</sub>. As a check on the detector sensitivity, a small ( $10^4$  n/s) <sup>252</sup>Cf source was

introduced at the same distance from the liquid-scintillation detector as the test cell. Count rates were 30 - 60 times background rates, and there was a notable shift in the spectrum of energies indicated by the MCA. There were no other indications of radioactivity other than background during the entire course of the experiments.

**Tritium Buildup.** Since heavy isotopes are electrolyzed more slowly, tritium tends to stay behind in solution. Assuming that electrolysis rate  $R_{el}$  and evaporation rate  $R_{ev}$  are roughly proportional to the inverse square root of the mass, the tritium fraction  $f$  in a volume  $V$  of heavy water will increase in time according to the equation

$$df/dt = f_s S/V - (f/V)(2/3)^{1/2}(R_{el} + R_{ev}), \quad [11]$$

where  $S$  is the rate at which new heavy water is added, containing a fraction  $f_s$  of tritium. The solution of this equation is

$$f/f_0 = \exp(-t/\tau) + (S\tau/V)(f_s/f_0)[1 - \exp(-t/\tau)], \quad [12]$$

where

$$\tau = (3/2)^{1/2}V/(R_{ev} + R_{el}) \quad [13]$$

and  $f_0$  is the initial value of  $f$ . For the INEL experiment  $S = R_{ev} + R_{el} = 3.0 \pm 0.2$  ml/day,  $R_{el} = 2.58 \pm 0.1$  ml/day,  $V = 40 \pm 0.2$  ml, and  $f_s = f_0$ . Therefore,  $\tau = 16.3 \pm 1.2$  days.

The initial tritium count rate from a 10 ml sample of our heavy water was  $880 \pm 8$  counts per minute. After 5.7 days of operation, Eq.[12] predicts  $f/f_0 = 1.066 \pm 0.004$ , from which the count rate is expected to be  $938 \pm 9$  counts per minute. The measured value at the end of 5.7 days was  $927 \pm 8$  counts per minute, in satisfactory agreement with the predicted value.

## CONCLUSIONS

The power balance residue,  $\Delta\dot{Q}$ , observed in the present experiments was effectively the same in  $D_2O$  experiments as in the  $H_2O$  control tests. The energy flows measured should be accurate to about  $\pm 5\%$ . Certainly, we did not see excess energies in multiples of input energies as reported by Fleischmann and Pons.<sup>1</sup> Evidence of radiation or residual tritium were lacking. We conclude that we did not observe evidence for fusion or any other nuclear process taking place in our experiments.

There are several possible explanations for perceived energy releases. Processes involving H/D interactions with the electrodes were evaluated (Appendix) and judged not to be significant. Irreversibilities associated with electrolysis, especially resistance and activation polarization effects, are much more important. One thing is very clear from these results: there are many irreversible processes going on which lead to a substantial heat flux from the test cell in comparison with the work of electrolysis.

## ACKNOWLEDGMENTS

We express grateful appreciation to Randy Shirts for his critical and enlightening consultations, to A. J. (Gus) Caffrey for assistance with neutron detection instruments and furnishing laboratory space, to Richard L. Miller for furnishing additional laboratory space and some of the Pt wire used for anodes. Also we are grateful to Stan Englert for his timely and expert technical assistance, and to the many others who assisted in varied ways with discussion, encouragement, and technical support. Finally, we thank the management at the INEL and EG&G Idaho, Inc. for making time available to pursue this work.



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TABLE 1. SUMMARY OF EXPERIMENTAL RESULTS

Cathode	Water	Operating Time (hr)	Applied Current (mA)	Potential (V)	Heat Transferred (mW)	Heat Increment (mW)
Coil	D <sub>2</sub> O	120	400	5.10	1,460	31
Coil	D <sub>2</sub> O	26	400	4.95	1,340	-25
Coil	D <sub>2</sub> O	5	200	3.56	448	33
Coil	Mixed	6	400	4.15	1,090	29
Rod	D <sub>2</sub> O	1	300	4.55	980	51
Foil	D <sub>2</sub> O	1	600	5.05	2,160	6
Foil	D <sub>2</sub> O	0.2	440	4.52	1,320	-31
Foil <sup>a</sup>	H <sub>2</sub> O	1.2	480	3.45	1,029	83
Foil <sup>b</sup>	H <sub>2</sub> O	0.6	480	3.51	982	7

<sup>a</sup> Cathode 80% immersed.<sup>b</sup> Cathode 60% immersed.

TABLE 2. CONSTANTS FOR THE VOLTAGE EQUATION FROM EXPERIMENTAL DATA

$$V = a + b \ln(I) + RI$$

Cathode	Water	LiOH Concentration	a (V)	b (V)	R (Ohm)
Coil	D <sub>2</sub> O	0.1 M	2.9	0.21	6.2
Coil	D <sub>2</sub> O	1.0 M	3.3	0.21	5.7
Coil	D <sub>2</sub> O	1.2 M	3.3	0.21	5.2
Coil	Mixed	0.1 M	2.7	0.11	6.2
Foil	D <sub>2</sub> O	0.1 M	4.2	0.32	6.0

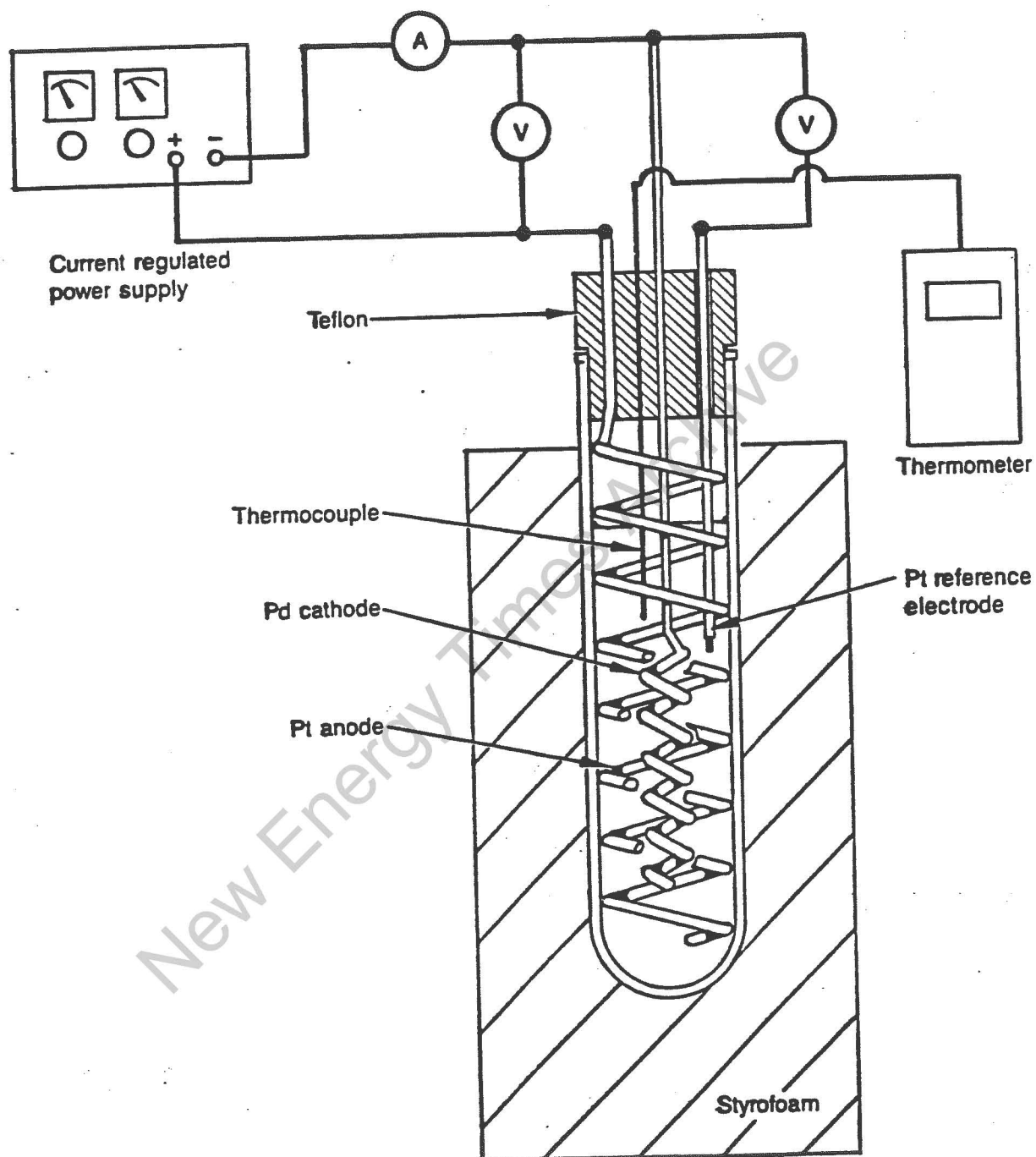


Figure 1. Schematic of the experimental apparatus used.

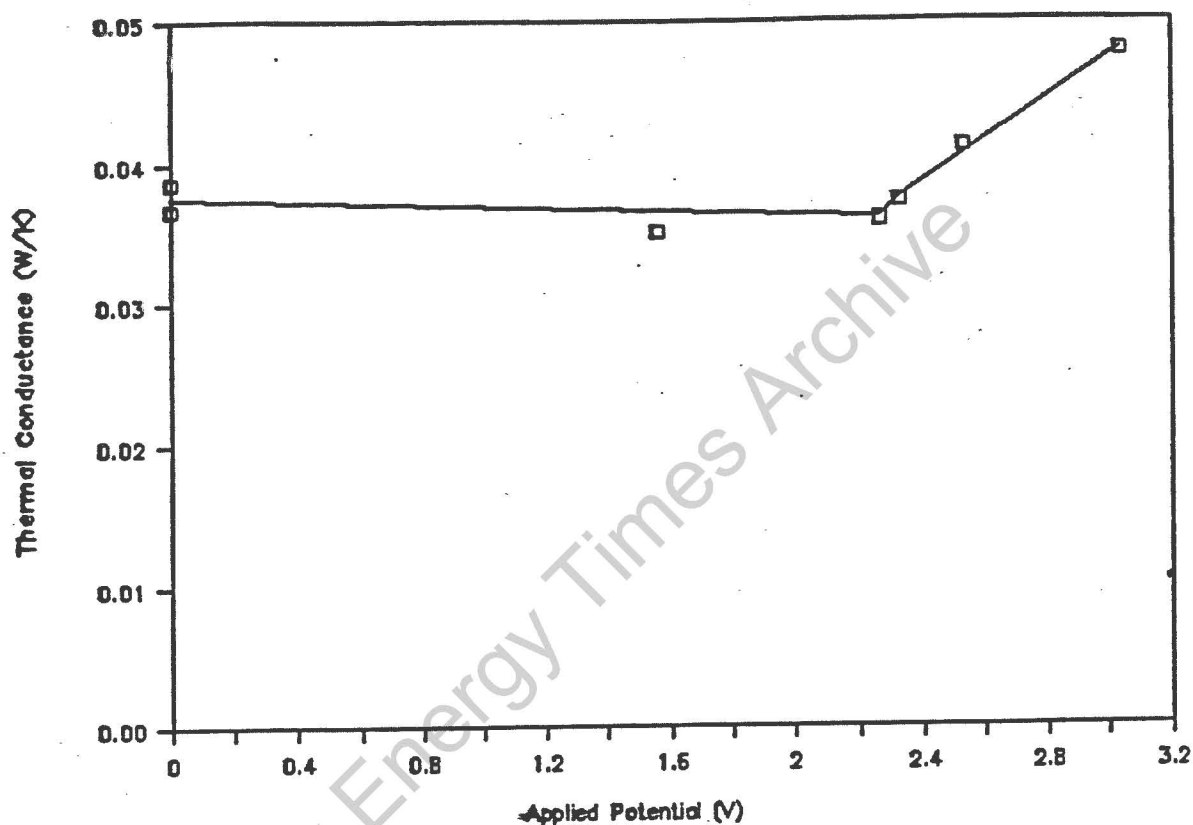
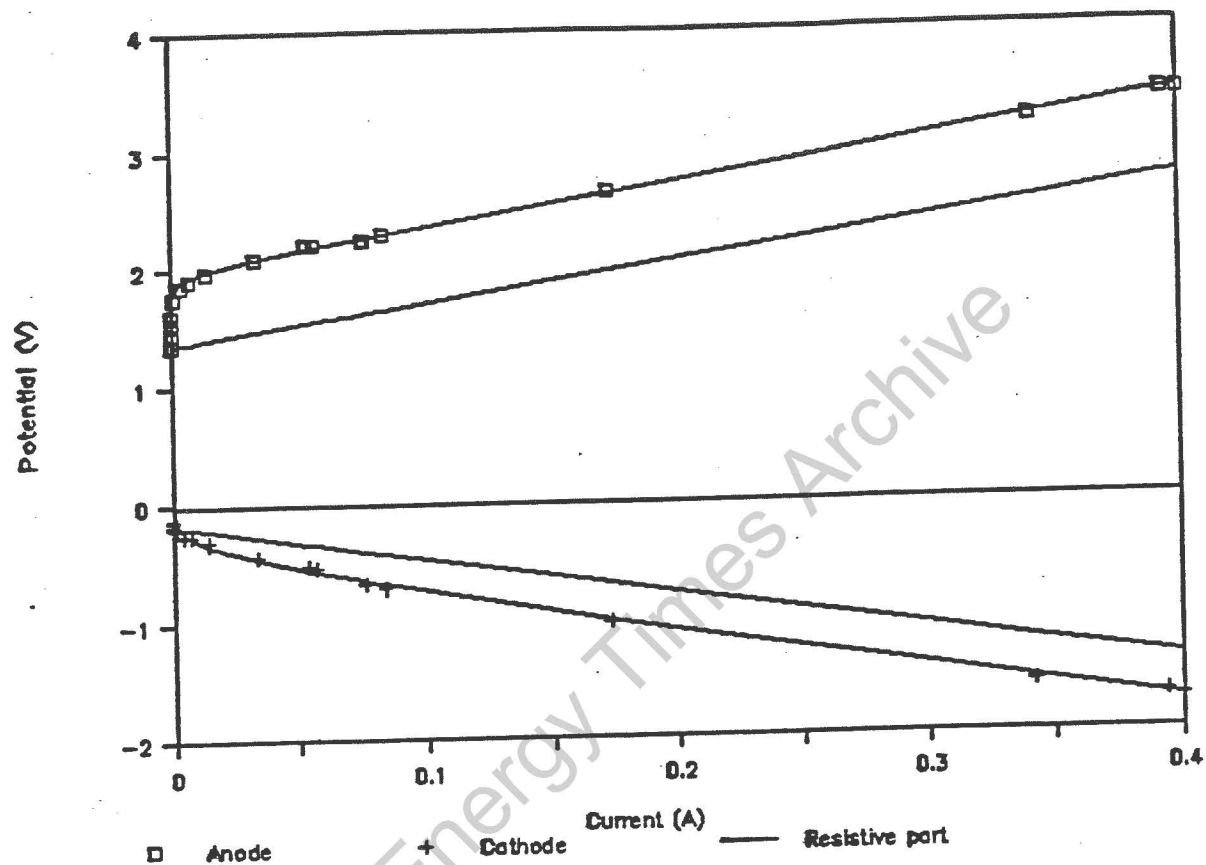


Figure 2. Thermal conductance measurements for electronic-resistor heated cells containing 30 mL of electrolyte. Above 2.2 V electrolysis was occurring in the cells.



**Figure 3.** Operating voltage characteristic curves for a coil cathode with 0.1 M LiOH showing the onset of activation polarization potentials at very low currents, and the relatively high impedance at higher currents. Lines drawn in are the resistance contribution to the total potential.

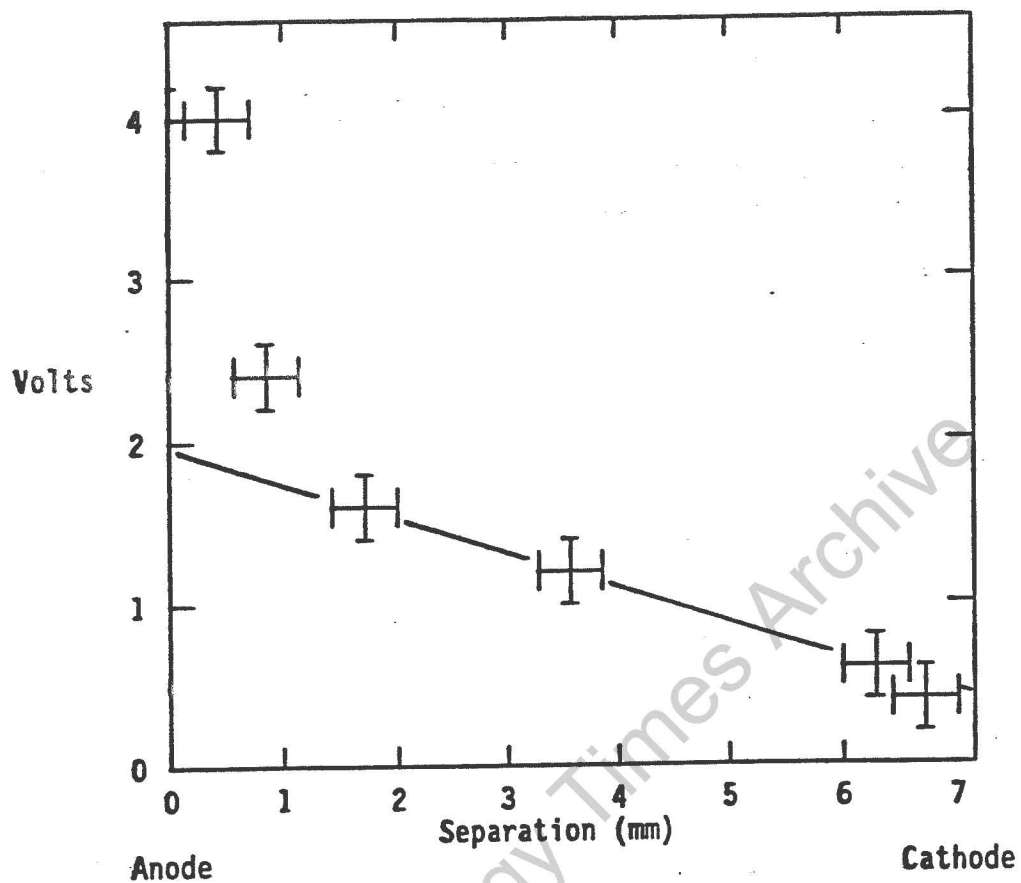


Figure 4. Potential variation between electrodes of a test cell operating at 200 mA as measured by a 0.5-mm diameter x 1-mm long Pt reference electrode.



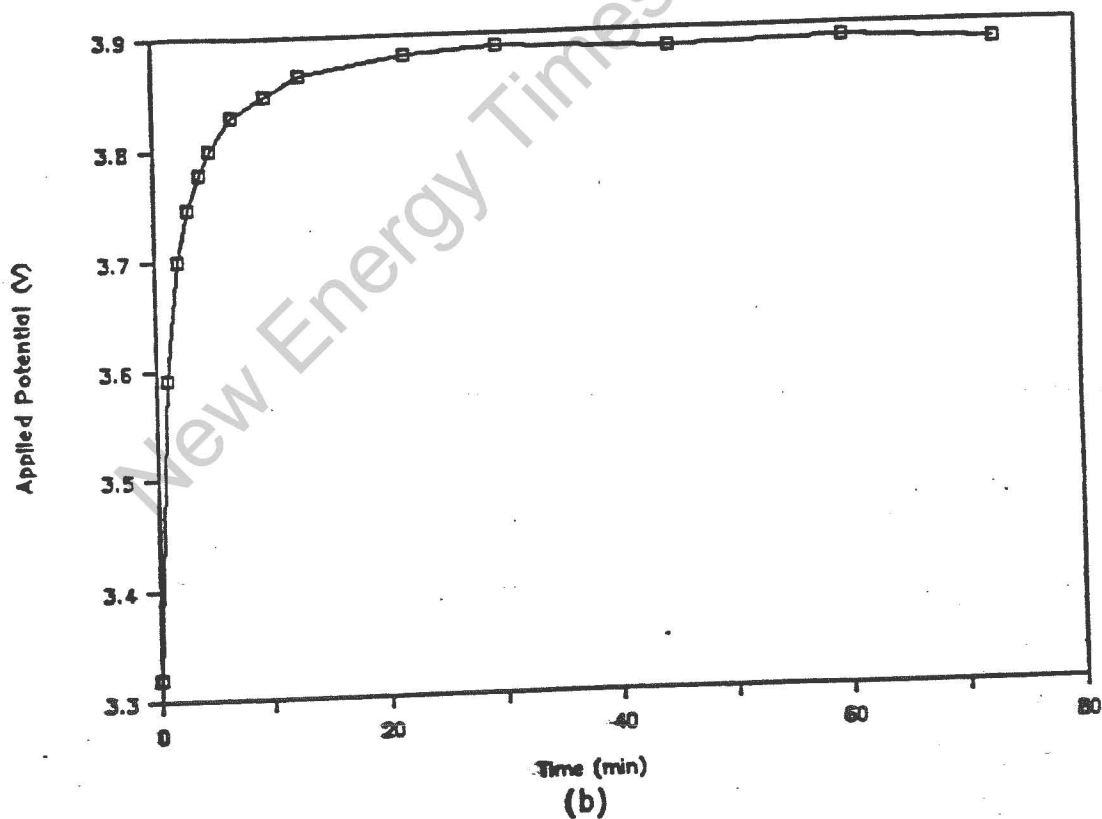
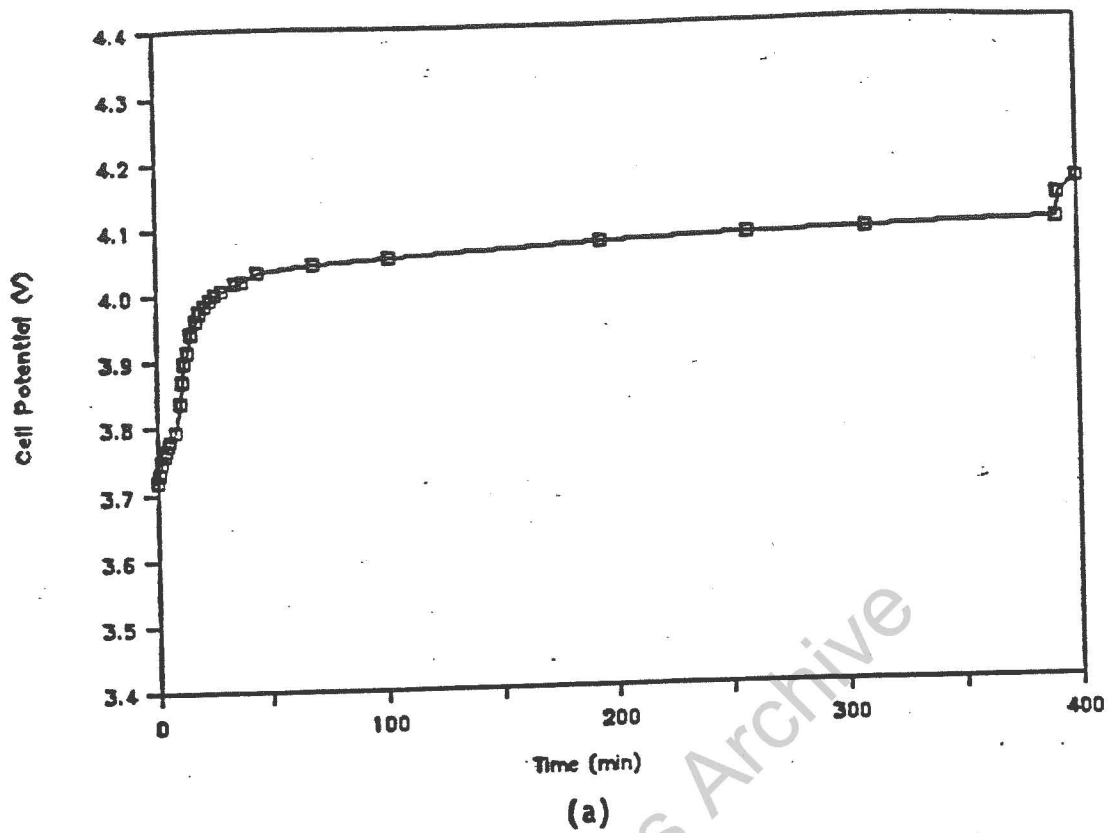


Figure 5. Voltage onset characteristics for (a) coil cathode, and (b) foil cathode. The voltage increment includes parts from surface effects and deuterium loading of the cathode.

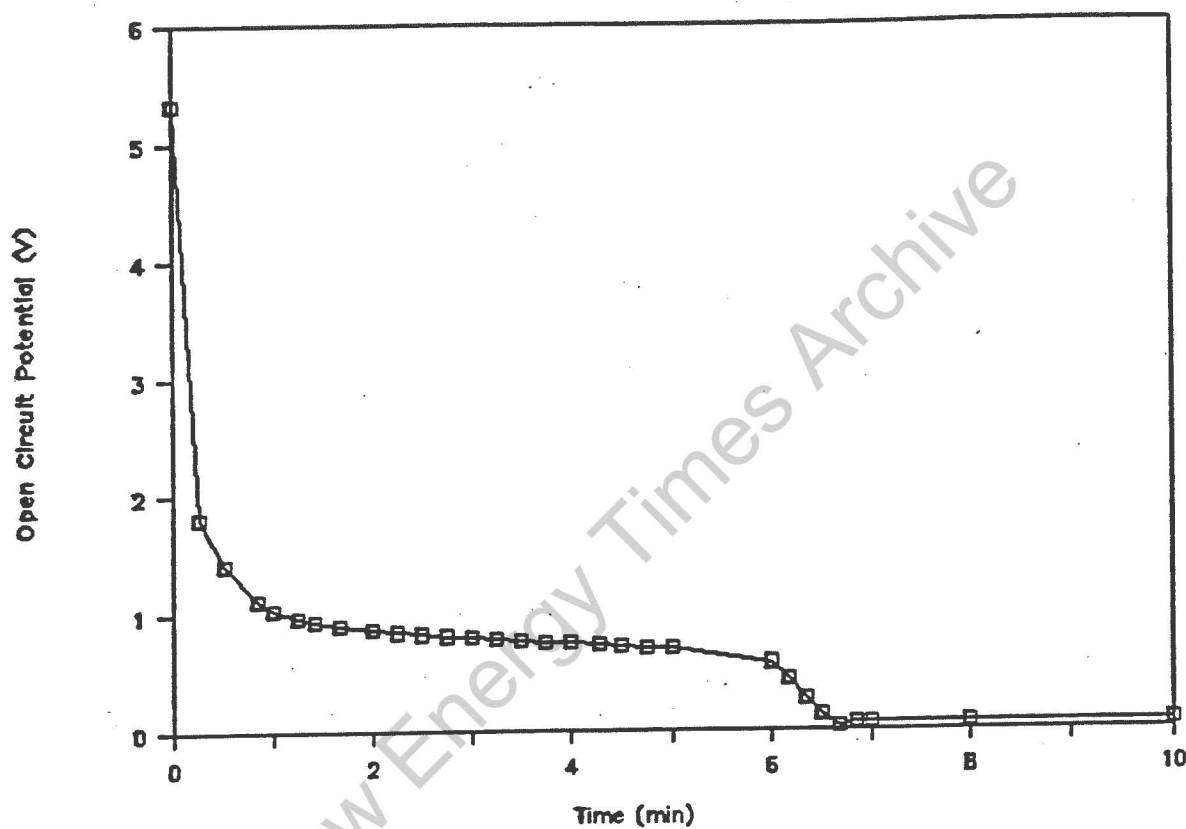


Figure 6. Open-circuit potential characteristic for coil cathode after 5.6 days of operation. This is believed to represent processes ongoing as the oxygen anode is transformed to a hydrogen electrode.

## APPENDIX

Water evaporation and several mechanisms involving hydrogen or deuterium interaction with the Pd cathode were evaluated to see if they may significantly alter the heat measurements in these experiments. Presented below are summaries of those considerations.

### EVAPORATION

Electrolysis products, hydrogen or deuterium ( $H_2/D_2$ ) and oxygen, leave the control volume (C.V.) and carry enthalpy with them. The molar flow rate for the hydrolysis of water in the C.V. is given by

$$\dot{n}_w = \frac{\eta I}{2F}$$

[A-1]

where the factor 2 is the result of 2 electrons being required per water molecule for electrolysis, and  $F$  is the Faraday constant, 96,485 coulombs per mole of electrons. The multiplier,  $\eta$ , is the fraction of the electrolyzed molecules that do not recombine into water before leaving the C.V. Measurements of water addition requirements over extended periods of cell operation show that  $\eta$  is very close to unity though some recombined water was observed on the Pt wire in the top of the cell. The volume occupied by the gas phase does not change substantially during the course of a measurement, hence it may be assumed that all of the gases electrolyzed leave the C.V. along with some water vapor. The  $H_2/D_2$  evolution rate is identical with  $\dot{n}_w$  while the oxygen evolution rate is half of that.

The small gas bubbles coming from the electrodes become saturated with water vapor at its saturation partial pressure,  $P_v$ , before emerging from the surface. The magnitude of the convective water loss rate and its associated energy flux may be evaluated as follows: If  $P_{amb}$  is the total ambient pressure, then

$$P_{amb} = P_{H_2} + P_{O_2} + P_v,$$

[A-2]

and by assuming ideal gas behavior it may be shown that the molar rate of entrained vapor leaving the cell is

$$\dot{n}_v = \frac{P_v}{P_{amb} - P_v} \frac{3\dot{n}_w}{2} \quad [A-3]$$

The electrolysis rate is  $\dot{n}_w$  (Eq.[A-1]), and at 298.15 K and 1 bar  $\Delta H(\text{elect})$  is 294.59 kJ/mole for  $D_2O$  (the negative of the water formation enthalpy change). The enthalpy change for  $D_2O$  evaporation at standard conditions is 45.4 kJ/mole. Assuming  $P_v$  of 0.03 bar, the molar ratio of water vapor is 4.6% (confirmed by measurement), the energy correction for evaporation is only 0.7%, and it may be neglected from further consideration.

### WATER REFORMATION

Pt and Pd are excellent catalysts for the oxidation of hydrogen. These materials are present in the space above the electrolyte. Oxygen and hydrogen or deuterium emerge from the surface of the electrolyte and mix in that space. It is conceivable that these metals could result in a significant fraction of the gas recombining to form water. If that happened, there would be an apparent heat excess because the flow term in Eq.[1] of the paper would be reduced.

That does not take place to any sizable extent as is evidenced by the requirements for water addition over extended periods of operation. Water droplets may be seen on the electrode leads after several hours. That apparently drowns the metal and prevents further catalysis. Further, the leads are small and relatively polished.

### ABSORBED HYDROGEN EFFECTS

The rate of change of enthalpy,  $\dot{H}$ , for the C.V. is the sum of the products of the change,  $\Delta H_i$ , for each phase transition taking place and the molar rate at which those changes take place, together with changes in  $H$  which may occur due to changes in temperature or pressure. The phase change considered important is mainly electrolysis of the water

to H<sub>2</sub>/D<sub>2</sub> and O<sub>2</sub>. Several possible interactions of hydrogen or deuterium (H/D) with the Pd cathode are now considered to evaluate the significance of their effects. Because the volume of the lead to the Pd was small and the Cu lead (used on some tests) has a very low solubility for H/D, uptake in the leads may be neglected.

### Temperature Change Effects

If the cell is operating at constant temperature and pressure, and no loading or unloading of the Pd with H/D is occurring, then only electrolysis need be considered. Because it may take a long time to load the cathode with H/D, and because temperature changes or metastable phases may occur that result in absorption or evolution of gas by the electrodes during a measurement, the magnitude of these effects should be evaluated.

An equation of state for hydrogen isotopes in the  $\beta$  phase of Pd is quoted by Wicke and Brodowski<sup>A-1</sup> as

$$\ln P^* = -\frac{\Delta H(x)}{RT} + 2 \ln \frac{x}{1-x} + \frac{\Delta S^0}{R} \quad [A-4]$$

where x is the H/Pd atom ratio and where the the enthalpy and entropy increments for the various hydrogen isotopes as they come out of solid solution are

	$\Delta H(x)$ (kJ/mole)	$\Delta S^0$ (J/mole.K)
H <sub>2</sub>	100.29 - 89.96 x	107.1
D <sub>2</sub>	95.48 - 89.96 x	106.3
T <sub>2</sub>	92.26 - 89.96 x	102.9

Taking P\* from Eq. [A-4] as that required in ambient gas to sustain atom ratio, x, at temperature T, the free energy change associated with a hydrogen isotope being absorbed in the Pd would be

$$\Delta G(s) = RT \ln P^* = -\Delta H + 2RT \ln \frac{x}{1-x} + T\Delta S^0, \quad [A-5]$$



The rate at which gas enters or leaves the Pd is governed by diffusion considerations and the surface potential.

Figure A-1 shows a calculation of Eq.[A-5] over a range of deuterium atom ratios,  $x$ , for several temperatures. Suppose the cell was operated for an extended period at an arbitrary but constant temperature and achieved an equilibrium value of  $x$  as determined by temperature and the effects of the electric field at the surface. Then suppose that by external means the temperature of the cell, including the Pd was raised. There would be suddenly an excess of deuterium in the Pd. That excess concentration would drive the gas from the metal. Because solution of H/D in Pd is exothermic, release of gas from the metal will have an associated thermal energy uptake. Conversely, a drop in cell temperature will cause more gas to be absorbed with an attendant release of thermal energy. The work done by the electric field in charging the Pd will depend on the H/D atom ratio,  $x$ . If  $\Delta G$  is positive, work must be done by the electric field to force the deuterium into solution creating a sink for electrical energy. Some of that will be converted to heat. At the beginning of a run, when the gas atom ratio,  $x$ , is low,  $\Delta G$  from ambient gas to solid solution is negative, and absorption will result in work being done on the surface electric field with an attendant liberation of electrical energy, some of which will result from the heat uptake.

To evaluate the energy flows as the deuterium enters the Pd, consider the case where  $\Delta G$  is positive in Figure A-1. The reversible work done by the electric field when deuterium goes into solution in an isothermal process would then be  $\Delta G$ , which for deuterium at 298 K and  $x = 0.95$  would be -36.09 kJ/mole (negative because work is being done on the deuterium). The enthalpy change for the deuterium on solution into Pd is  $\Delta H$  or -10.0 kJ/mole (note that the values listed following Eq.[A-4] are for the evolution reaction).. The heat transferred would be  $-T\Delta S$  or 46.11 kJ/mole (thermal energy comes from the deuterium). The process is reversible, so the opposite transfers occur when the gas diffuses out of solution in the presence of the surface electric field.

The rate of gas release would be governed by diffusion considerations and would drop as equilibrium is approached. The duration of the gas



release would be effectively the diffusion time of deuterium in the Pd cathode. For the 1-mm diameter wires such as used in these experiments, the diffusion time is about 7 hours, and the wires contain about 0.012 moles of Pd. From Figure A-1 the change in  $x$  for a 20 K temperature change is about 0.01. If 0.00012 moles of gas were to come out over a 7-hour period, the thermal power level would be 0.22 mW (absorbed). That would be in the noise level in our experiments. If the cathode was 4 mm in diameter instead, the number of moles it contained would increase by a factor of 16. However, the diffusion time would also increase by the same factor, so the power level seen should remain the same.

### Gas Pumping

Suppose the cathode was only partly immersed in electrolyte. If it had a significant cross-sectional area, gas forced into it by the electric field at its interface with the electrolyte would diffuse up through the plane of the electrolyte surface and out into the free space around the cathode above the electrolyte. As the gas left the electrode in the absence of the confining electric field, there would be an absorption of thermal energy to accommodate the increase in enthalpy. It would result in a cooling of the electrode and the gases surrounding it. The entropy increase attendant to its release would be generated by the effectively unconstrained expansion of gas as it left. If those gases left the C.V. at a lower temperature than the bulk C.V. temperature, the result would be manifest as an added heat transfer to the surroundings (heat released as the gas went into solution that does not appear as  $h$  in the flow term of Eq.[1] in the paper). This would be a highly irreversible process that would increase in potency as the cross section of the electrode increased.

The magnitude of the heat would depend on geometry and on  $x$  because at high  $x$  the work to get the deuterium into the lattice is greater. Thermal equilibration between the cooler gas and the warmer liquid would also be an important factor. To give a rough estimate, suppose the diffusional gas flux was characterized by a gradient of  $1.4 \times 10^{24}$  D/cm<sup>4</sup> ( $x$  going from .95 to 0 over the 0.5-mm radius of the rod), a

diffusivity of  $2 \times 10^{-7} \text{ cm}^2/\text{s}$ , and a rod cross sectional area of  $7.8 \times 10^{-3} \text{ cm}^2$ . The diffusional flux would be  $3.6 \times 10^{-9} \text{ mole/s}$ . At 46.1 kJ/mole (the uncompensated heat release as the gas went into the Pd, assuming no heat exchange between the liquid and gas phases) the incremental heat transfer would be 0.16 mW. Again, this would be inconsequential in our experiments. It would scale linearly with cathode radius.

## THERMOELECTRIC EFFECTS

Peltier and Thomson heats were considered as possible contributors to heat flows, but it was concluded they would be unimportant. Table A-1 lists Peltier coefficients derived from the data of Ref. A-2 for the various junctions present in our test cells using the relation between Seebeck and Peltier coefficients,

$$\Pi_{AB} = T(S_B - S_A).$$

[A-6]

It may be seen that for  $\Pi_{AB}$  on the order of a few millivolts, the fractional ampere currents used in our experiments would mean sub-milliwatt power levels. These may be observable but will not substantially alter the outcome of the experiments.

TABLE A-1. DERIVED PELTIER COEFFICIENTS FOR SELECTED JUNCTIONS

Material	Thomson Coefficient ( $\mu\text{V/K}$ )	Seebeck Coefficient ( $\mu\text{V/K}$ ) <sup>a</sup>
Cu	1.2	1.5
Pd	-10.1	-10.0
Pt	-7.8	-5.5
Junction	Peltier Coefficient (mV)	
Cu-Pd	-3.45	
Pd-Pt	1.35	
Pt-Cu	2.10	

<sup>a</sup>Data taken from Ref. A-2.

Thomson heat is given by

$$\dot{Q}_T = \kappa_T I \cdot VT$$

[A-7]

where  $\kappa_T$  is the Thomson coefficient and  $I$  is the current in the conductor. It is taken positive if heat is liberated reversibly when  $I$  and  $\nabla T$  are antiparallel. If the same material is used for leads going from  $T_1$  at the cell boundary to a power supply which may be at  $T_2 < T_1$ , there should be no net heating effect due to Thomson heat. If the leads are of different materials, the heat gained by one will be different from the heat lost by the other, the difference appearing as a perturbation to the energy balance of Eq.[1]. Suppose that  $T_1 - T_2$  was 60 K and that a current of 500 mA was being drawn through Pd and Pt leads. A simple calculation shows that the incremental heat would be 76  $\mu$ W. Again, this is negligible.

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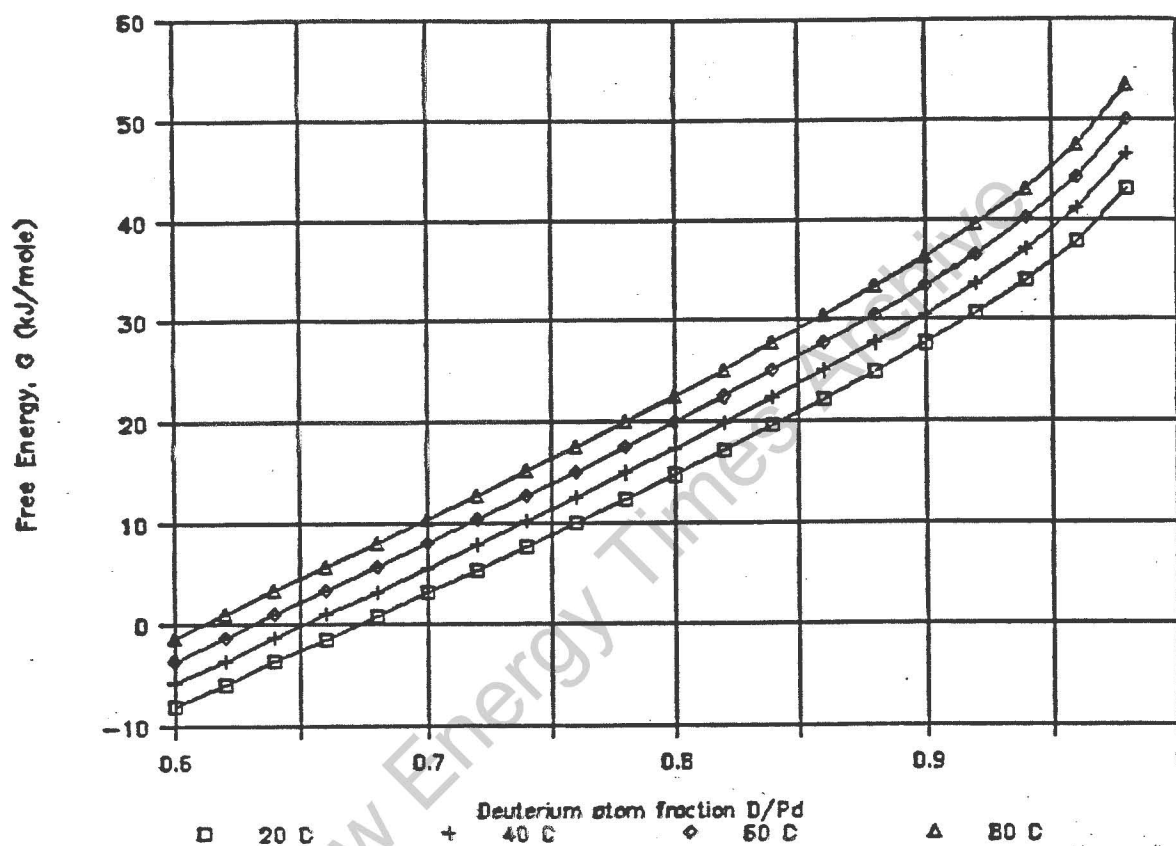


Figure A-1. Calculated free energy shifts for solution of deuterium in Pd at various solute atom ratios,  $x$ , and temperatures of 20, 40, 60, and 80 C.





September 6, 1989

Dr. John R. Huizenga  
Co-Chairman, ERAB  
Panel on Cold Fusion  
Energy Research Advisory Board  
U.S. Department of Energy  
1000 Independence Avenue S.W.  
Washington D.C. 20585

Dear Dr. Huizenga:

Here is the reply to your letter of August 9th. The answers match the numbers in your questionnaire.

1. Cambridge Isotopes Laboratory  
Woburn, MA  
99.9% purity  
Reported analysis data vary from 13 to 24 DPM.
2. Cell description:  
See attached figure  
100 to 120 ml of electrolyte.
3. Cell operating conditions:  
Earlier cell operations were in constant voltage mode.  
Approximate current densities were around 100 ma/cm<sup>2</sup>. Cell operation since second week of May is constant current.

Current mode: LiOD concentration - 0.1 molar nominal  
Open cells: D<sub>2</sub>, O<sub>2</sub> vented out  
Anode winding: platinum  
Cathode: - 4 mm palladium dia x 10 cm length  
Surface treatment: - various treatments given to the electrodes

4. D<sub>2</sub>O additions to match the current through the cell (within 1%).  
Approximately 8-10 cc/day.  
Earlier: added 3 times a day.  
Now: added continuously with syringe pumps.
5. Tritium analysis:
  - a) Results on our samples are being reported to you separately by Dr. John Morrey of Batelle Northwest.
  - b) Liquid scintillation detectors
  - c) Cells were examined after at least one heat excursion

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had occurred. One sample each of the initial  $D_2O$  + LiOD, was submitted along with the above samples.

Tritium analyses for electrolyte from one cell showing excess heat generation. (Analyses reported to us by Dr. John Morrey.)

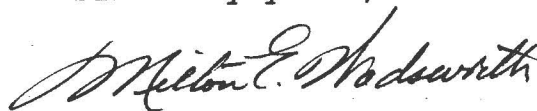
Sample #	Pd Rod #	Days of Operation	% of electrolyte exchange	Tritium atoms/ml
Blank $D_2O$	--	--	--	$2.12 \pm 0.70 \times 10^8$
Blank $D_2O + LiOD$	--	--	--	$1.32 \pm 0.71 \times 10^8$
3-1	#2	17	142	$9.4 \pm 0.81 \times 10^8$
3	#2	26	218	$9.85 \pm 0.82 \times 10^8$

6. Low level measurements not attempted.

7. The electrode #3 referred to in the above table produced a heat excursion lasting 90 minutes. The temperature rise in the cell was from  $31^\circ C$  to  $47^\circ C$  peak temperature which then dropped to  $42^\circ C$  where it remained. The entire episode lasted 90 minutes. The input voltage and currents were 9.76 volts and 0.95 amperes. The input power was 9.3 watts and the average power output was 44 watts. The total heat generated during the excursion was 187,000 joules. Samples 3-1 and 3 above were taken after the observed heat excursion.

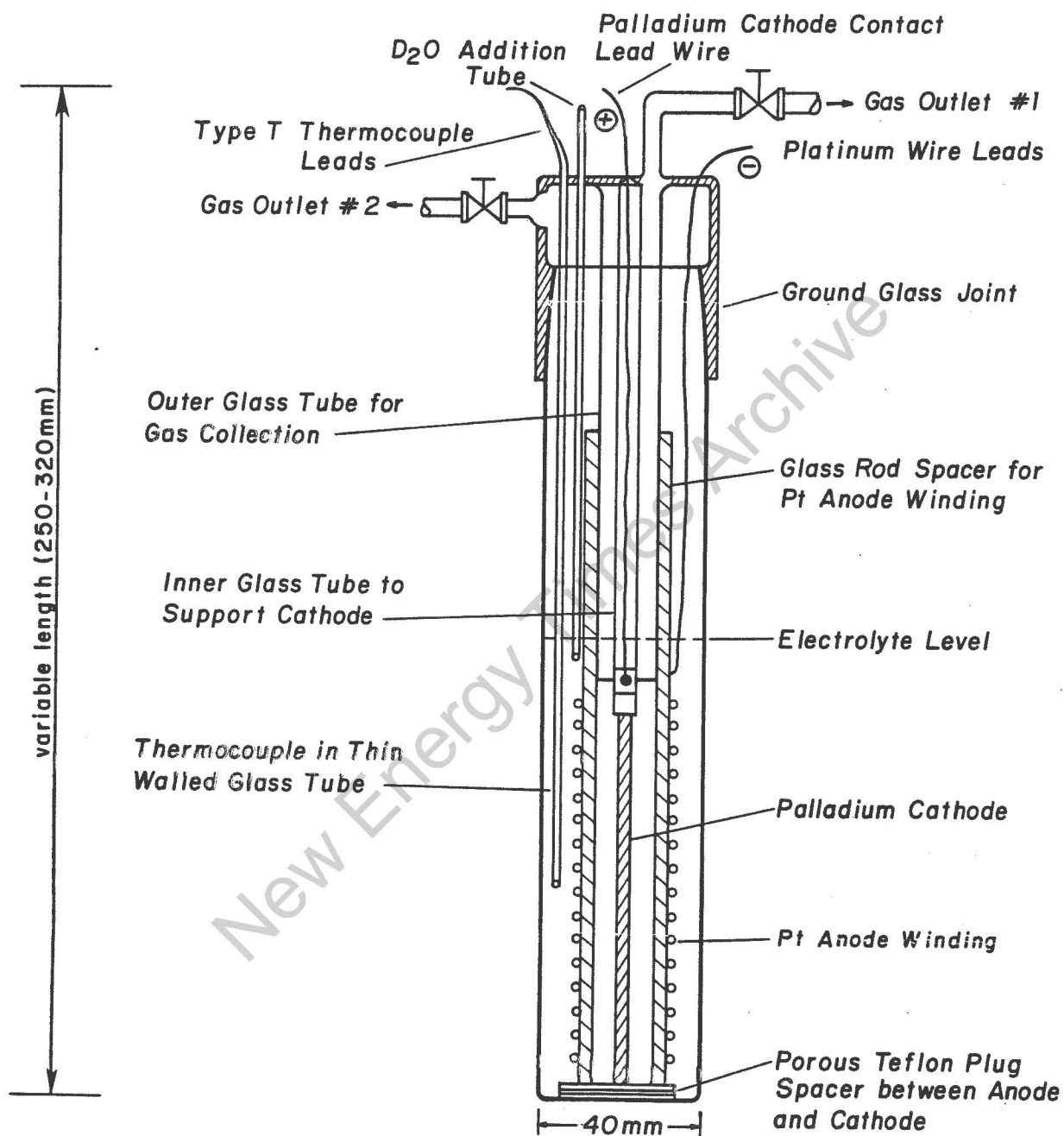
8. None in this building

Sincerely yours,



MILTON E. WADSWORTH  
Professor of Metallurgy  
and Dean





Dr. William Woodrad  
Secretary , Cold Fusion  
ER-6 , 3F-043  
US Department of Energy

28 August 1989

Dear Dr. Woodrad:

I just received a letter from Prof. Hora and Prof. Scheid.  
They recommend to send to you information about the cold  
fusion experiments which we have done at our institute.  
So I enclose two papers, the internal report about our  
first experiment and a copy of the article which will be  
published in Proceedings of the ICENES'89.  
I hope they will be useful to you.

Sincerely Yours

Jozef Farny

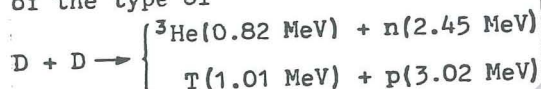
# INVESTIGATIONS OF NEUTRON EMISSION IN A COLD FUSION EXPERIMENT IN PALLADIUM

M. Szustakowski, J. Farny, M. Muniak, A. Nowak, P. Parys, W. Skrzeczanowski,  
R. Socha, J. Teter, J. Wolski, J. Wołowski, E. Woryna

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## 1. INTRODUCTION

The experiments dealing with performance of nuclear fusion at room temperature actually create a great sensation and are carried out in various laboratories [1,2]. This interest arises from the results achieved by Fleischmann and Pons, and it results from their paper [1] that there exists a possibility of obtaining an ignition owing to nuclear fusion reactions during "usual" electrochemical process - namely the electrolysis of D<sub>2</sub>O with use of the system of Pd-Pt electrodes. These fusion reactions of the type of



are connected with neutron emission. From this reason the measurements of the yield and behaviour of neutron emission give the information about processes of interest.

At the IPPLM the cold fusion experiments have been conducted from the beginning of April 1989. In the first experiment the reliable evidence of neutron emission was obtained. A number of irregularly repeated neutron pulses of the level of 10<sup>5</sup> per pulse was recorded. The measurements of the neutron emission, in this experiment, were performed with the use of three independent methods employing the 2.5 MeV neutron spectrometer, the scintillation neutron detector as well as the nuclear track detector. Neutron emission had been first recorded after 106 hours of the electrolysis process of D<sub>2</sub>O.

The correlation between the neutron emission and measured temperature effects has not been observed. The recorded electrode temperature changes

were clearly connected with the polarization current density.

In the next our experiments, besides diagnostic methods listed above, the silver activation neutron detectors and the triple coincidence system have been employed. In these experiments a disagreement, between results obtained by means of the nuclear track detectors and other diagnostic systems, appeared. This disagreement will be discussed in the next sections.

## 2. EXPERIMENTAL ARRANGEMENT

### 2.1. Experimental conditions

The scheme of the experimental arrangement used in the first experiment is shown in Fig. 1. The glass vessel, in which the electrolysis had been driven, was in cylinder shape 90 mm in diameter and 200 mm in height. The 0.5N NaOD solution in D<sub>2</sub>O (99.8% enhancement) as an electrolyte was used. Palladium electrode (a massive cylindrical rod cathode) was 10 mm in diameter and 124 mm in height, while the platinum grid of the 35 mm in diameter and 100 mm in height was used as an anode. During the first experiment the palladium electrode temperature was being continuously checked by means of a thermocouple.

During the first experiment the current density was changed from 12.5 to 105 mA/cm<sup>2</sup>.

In the next experiments similar geometry of electrodes and current densities were employed with one exception in which electrodes in the form of cylinders were used. In the last experiment the solution of <sup>6</sup>LiOD in D<sub>2</sub>O was used as the electrolyte.

End of text

## 2.2. Neutron emission measurement

A detailed scheme of the neutron measurement systems used scintillation probes in the first experiment is shown in Fig. 2.

The probe named NS consisted of the NE-213 liquid scintillator and PM tube. This probe was connected to the pulses counter via n- $\gamma$  discriminator, so only pulses from the neutron recoil protons could be counted. The overall efficiency of the NS probe (including measurement geometry) was equal to 0.3%.

The probe named NC consisted of the NE-102A plastic scintillator and PM tube and was connected to the pulses counter via amplitude discriminator only, so this probe was able to count pulses from neutron recoil protons pulses as well as the  $\gamma$ -rays from a reaction



The overall efficiency of the NC probe was 3.5%, especially because of the plastic scintillator volume which was about six times larger than the liquid one. In this case, the neutrons could be slowed down (in the scintillator) to energies at which the n-p reaction became effective.

The time gate of NS probe operating was 500 s (each measurement was carried out in a time interval equal to 500 s). The time gates for measurements with use of NC detector had been chosen to 50, 40 and 20 s, respectively, and for time gates of 40 and 20 s discriminator levels were increased.

The plastic nuclear track detectors (CR-39 type) were placed close to the wall of the glass vessel. Before experiments they were calibrated with use of Am-Be neutron source as well as by means of the "plasma focus" discharge, in which the short (about 100 ns) neutron pulses were emitted (about  $10^9$  neutrons in one pulse).

The neutron registration efficiency of these track detectors,  $\eta$ , determined by these methods amounted to  $2 \times 10^{-3}$ . The CR-39 track detectors were employed in all our experiments.

The scheme of neutron measurement system used in the next experiments is shown in Fig. 3. This system consisted of three scintillation probes NC, N3 and SND with the NE-102A scintillators. Scintillation probes were connected to a triple coincidence system and, parallelly, to counters. The probe named SND was connected also to an oscilloscope in order to mea-

sure the time duration of neutron pulses.

Aside from scintillation probes the two silver activation neutron detectors were used in this series of experiments. The neutron registration efficiency  $\eta$  of these detectors was about  $10^{-5}$  to  $10^{-6}$ , respectively.

## 3. RESULTS AND DISCUSSION

All the neutron emission acts registered during the first experiment are presented in Fig. 4. The main feature of the neutron emission in our experiment was its pulsed character. The time duration of the single neutron pulse was much shorter than 1 s. The main part of neutron pulses was recorded only by one (NC) detector. Only in two cases the neutron pulses were recorded by two detectors (NS and NC) simultaneously. This can be attributed to the following facts:

- the NC probe had the overall efficiency about one order of magnitude greater than the NS,
- the NC probe could register the neutron recoil protons and  $\gamma$ -ray from the n-p reactions "simultaneously", but the NS probe could not, because it had the n- $\gamma$  discriminator,
- if the time duration of the neutrons pulse had been shorter than about 10  $\mu$ s, then, because of the dead time of the NS probe electronic system, the neutron pulse was not registered and/or it was counted as a background.

The absolute neutron yield measured in this experiment, by track detector ( $5 \times 10^9$ ) neutrons in compare with neutron yield registered in one pulsed discharge recorded by NC and/or NS detectors show multiple emission of very short ( $\Delta t < 10$  ms) neutron pulses. However, our detectors were not optimized for registration of such short and intense neutron radiation pulses, and probably were not able to record a significant part of them.

Unfortunately, in our experiments such clear evidence of the pulsed neutron emission was not obtained. Furthermore, the measurements results had obtained by means of neutron spectrometer provided the evidence that there was not any continuous neutron emission. However, in all our experiments the track detector results indicated the total neutron yield on the level of  $3 \times 10^8$  -  $5 \times 10^9$ . So there is a serious disagreement between the track detectors results and those obtained by other diagnostics (scintillation probes, silver activation detectors).

*$10^5$  -  $10^6$  neutrons  
in one pulse*

Fig. 3. Neutron measurement system. Size 104 x 71 inches

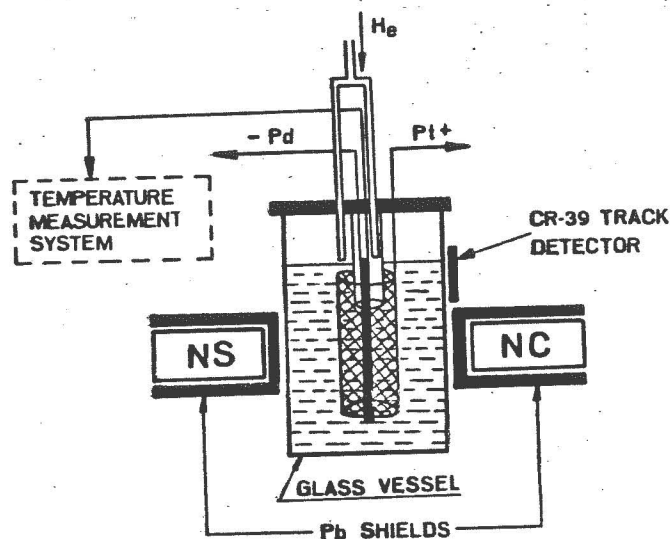


Fig. 1. The scheme of the experimental arrangement: NS - 2.5 MeV neutron spectrometer, NC - scintillation neutron counter, electrolyte - 0.5N NaOD solution in D<sub>2</sub>O (99.8%)

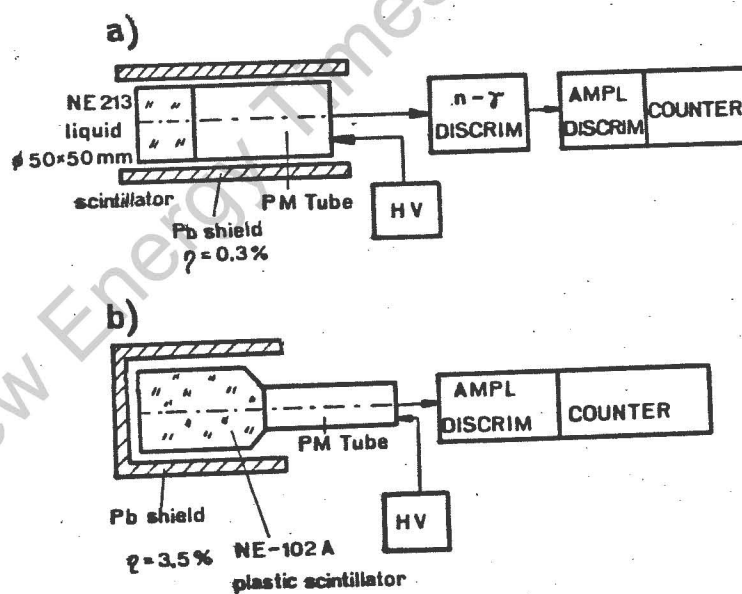
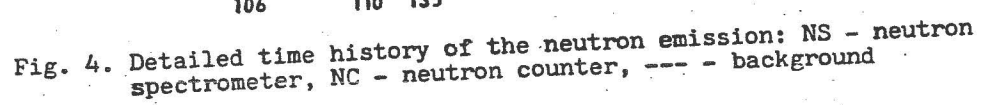
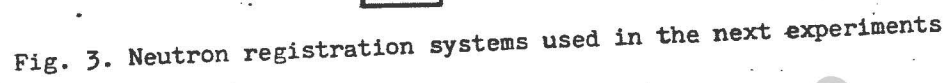


Fig. 2. Neutron registration systems used in the first experiment: a - 2.5 MeV neutron spectrometer, b - scintillation neutron counter, η - total registration efficiency (including the geometry of measurements)







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In the near future experiments the authors will try to explain these disagreements which, maybe are connected with the specific time and space character of neutron emission.

We wish to thank W. Alifanow, H. Włodarczyk and W. Trociński for their help and assistance during this experiment.

REFERENCES: text below this line

1. M. Fleischmann, S. Pons, Electrochemically induced nuclear fusion of deuterium, Journal of Electroanalytical Chemistry, March, 1989.
2. S.E. Jones et al. and J. Rafelski, Observation of cold nuclear fusion in condensed matter, March, 1989.

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PALLADIUM**

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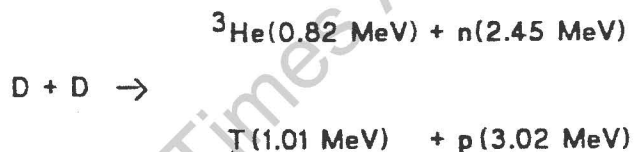
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# THE EVIDENCE OF NEUTRON EMISSION IN A COLD FUSION EXPERIMENT IN PALLADIUM

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J.Teter, J.Wolski, J.Wołowski, E.Woryna.

## 1. Introduction

The experiments dealing with performance of nuclear fusion at room temperature actually create a great sensation and are carried out in various laboratories [1,2]. This interest arises from the results achieved by Fleischmann and Pons, and it results from their paper [1] that there exists a possibility of obtaining an ignition owing to nuclear fusion reactions during "usual" electrochemical process - namely the electrolysis of  $D_2O$  with use of the system of Pd-Pt electrodes. These fusion reactions of the type of



are connected with neutron emission. From this reason the measurements of the yield and behaviour of neutron emission give the information about processes of interest.

In 1989, April the 14th, during the experiment carried out at IPPLM in Warsaw, according to the idea proposed by Fleischmann and Pons the nuclear fusion events at room temperature were registered. The reliable evidence of neutron emission was obtained. The measurements of the neutron emission were performed with the use of the three independent methods employing 2.5 MeV neutron spectrometer, the scintillation neutron detector as well as the track detector. A number of irregularly repeated neutron pulses of the level of  $10^6$  per pulse was recorded. This important fact, reported in the literature for the first time, may have the crucial meaning for explaining the physical processes occurring during this phenomenon. Neutron emission had been first recorded after 106 hours of the electrolysis process of  $D_2O$ .

Up to now the correlation between the neutron emission and measured temperature effects has not been observed. The recorded temperature

changes were clearly connected with the polarization current density. Actually the interpretation of the observed processes is undertaken.

## 2. Experimental arrangement

### 2.1. Experimental conditions

The scheme of the experimental arrangement is shown in Fig.1. The glass vessel, in which the electrolysis had been driven, was in cylinder shape 90 mm in diameter and 200 mm in height. The 0.5N NaOD solution in D<sub>2</sub>O (99.8 % enhancement) as a electrolyte was used. Palladium electrode (a massive cylindrical rod cathode) was 10 mm in diameter and 124 mm in height, while the platinum grid of the 35 mm in diameter and 100 mm in height was used as an anode. During the whole experiment the palladium electrode temperature was being continuously checked by means of a thermocouple. For the neutron registration the two scintillation detectors called as NS and NC were used. The CR-39 nuclear track detector was used for the measurement of the total number of emitted neutrons.

### 2.2. Neutron emission measurement

A detailed scheme of the neutron measurement systems using scintillation NS and NC probes is shown in Fig. 2.

The probe called NS consisted of the liquid scintillator of the NE-213 type and PM tube. This probe was connected to the pulses counter via n- $\gamma$  discriminator, so only pulses from the neutron recoil protons could be counted. The overall efficiency of the NS probe (including measurement geometry) was equal to 0.3%.

The probe called NC consisted of the NE-102A plastic scintillator and PM tube and was connected to the pulse counter via amplitude discriminator only, so this probe was able to count pulses from neutron recoil protons pulses as well as the  $\gamma$ -rays from a reaction:



The overall efficiency of the NC probe was 3.5%, especially because of the plastic scintillator volume which was about six times larger than the liquid one. In this case the neutrons could be slowed down (in the scintillator) to energies at which the n-p reaction became effective.

The time gate of NS probe operating was 500 s ( each measurement was

carried out in a time interval equal to 500 s). The time gates for measurements with use of NC detector had been chosen to 50, 40 and 20 s, respectively, and for time gates of 40 and 20 s discriminator levels were increased.

The geometry of the measurement with use of track detector (CR-39 type) is shown in Fig. 3. These track detector had been calibrated against Am-Be neutron source before the experiment. Chemical etching of track detector was carried out in 6.25N NaOH solution at temperature of 70 °C for 1 hour and after "developing" the tracks were observed and counted with use of optical microscope.

### 3. Results and discussion

The time history of the whole process, including time changes of polarization current  $I$ , electrode temperature  $T$  as well as some registered neutron pulses is shown in Fig. 4. A clear correlation between polarization current and electrode temperature is seen. Such a correlation between the neutron emission and the electrode temperature has not been observed.

All the neutron emission acts registered by scintillation detectors are presented in Fig. 5. The main feature of the neutron emission in our experiment was its pulsed character. The time duration of the single neutron pulse was much shorter than 1 s. The main part of neutron pulses was recorded only by one (NC) detector. Only in two cases the neutron pulse was recorded by two detectors (NS and NC) simultaneously. This can be attributed to the following facts:

- the NC probe had the overall efficiency about one order of magnitude greater than the NS,
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- if the time duration of the neutrons pulse had been shorter than about 10  $\mu$ s, then, because of the dead time of the NS probe electronic system, the neutron pulse was not registered and/or it was counted as a background.

The results obtained with use of track detector show the neutron flux at the level of  $10^5$  n/s, if the assumption on continuous emission, [1], is made.

Additionally, if one recalculates our Pd electrode volume the result is similar as Fleischmann and Pons one, [1]. However, the measurements performed with use of scintillator probes clearly show the pulsed character of neutron emission. Moreover, absolute neutron yield measured by track detector ( $\sim 10^{10}$  neutrons) in compare with neutron yield registered in one pulsed discharge recorded by NC and/or NS detectors show multiple emission of very short ( $\Delta t < 10$  ms) neutron pulses. However, our detectors were not optimized for registration of such short and intense neutron radiation pulses, and probably were not able to record a significant part of them.

In near future experiments the autors will try to estimate the time duration of neutron pulses as well as the connection between neutron generation and parameters of electrolysis process.

We wish to thank W.Alifanow, A.Włodarczyk and W.Trociński for their help and asistance during this experiment.

#### References

1. M. Fleischmann, S. Pons, Electrochemically induced nuclear fusion of deuterium, Journal of Electroanalytical Chemistry, March, 1989.
2. S. E. Jones et al. and J. Rafelski, Observation of cold nuclear fusion in condensed matter, . March, 1989.



## FIGURE CAPTIONS

Fig.1. The scheme of the experimental arrangement:

NS - 2.5 MeV neutron spectrometer,

NC - scintillation neutron counter,

E - 0.5 N NaOD solution in  $D_2O$  (99.8% enhancement)

Fig.2. Neutron registration systems:

A - 2.5 MeV neutron spectrometer,

B - scintillation neutron counter,

$n_t$  - total registration efficiency

Fig.3. Time history of the experiment:

I - polarization current,

T - electrode temperature,

N - number of counts of neutron emission

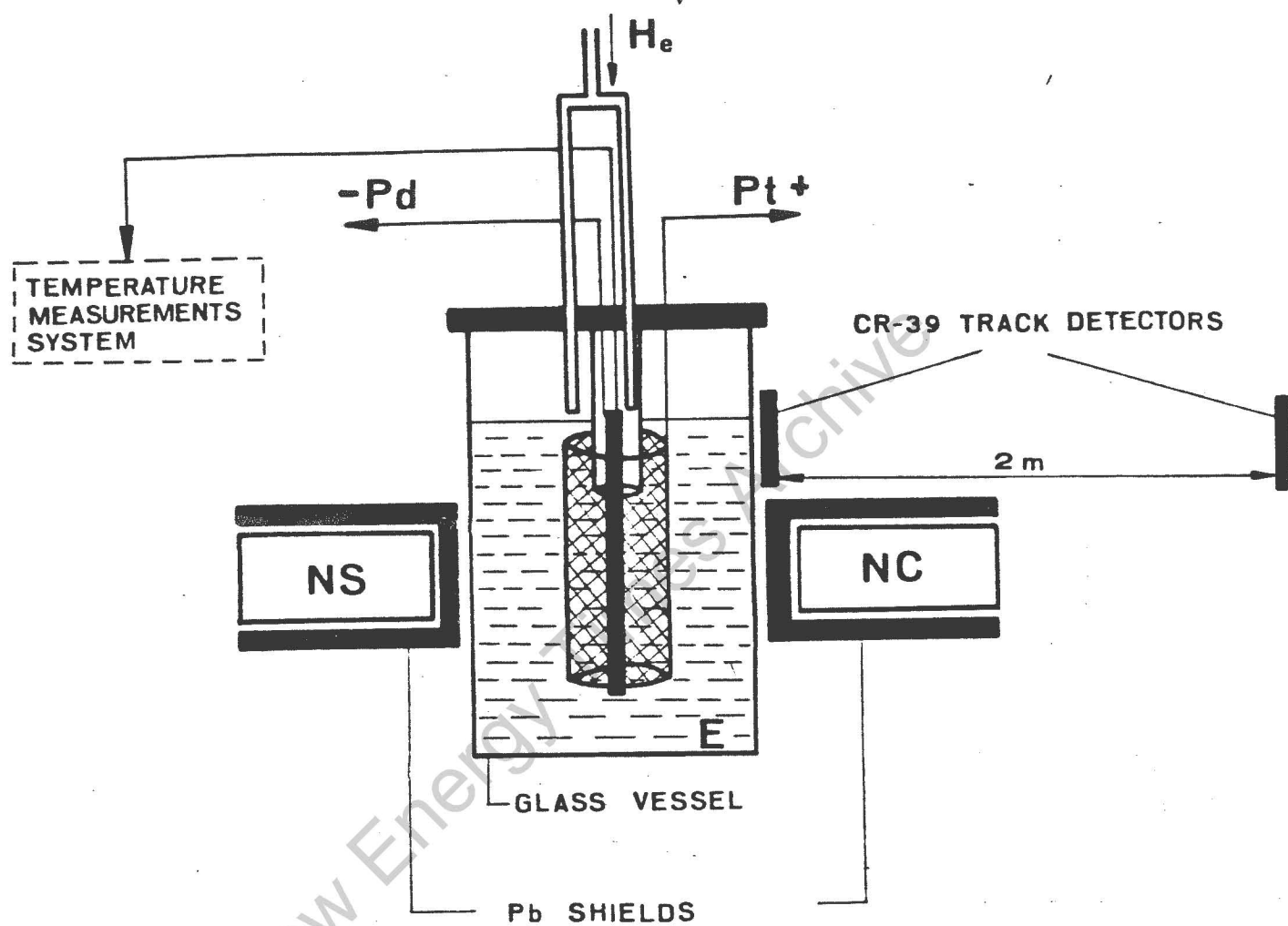
Fig.4. The geometry of CR-39 track detector measurements

Fig.5. Detailed time history of the neutron emission:

NS - neutron spectrometer,

NC - neutron counter,

---- background



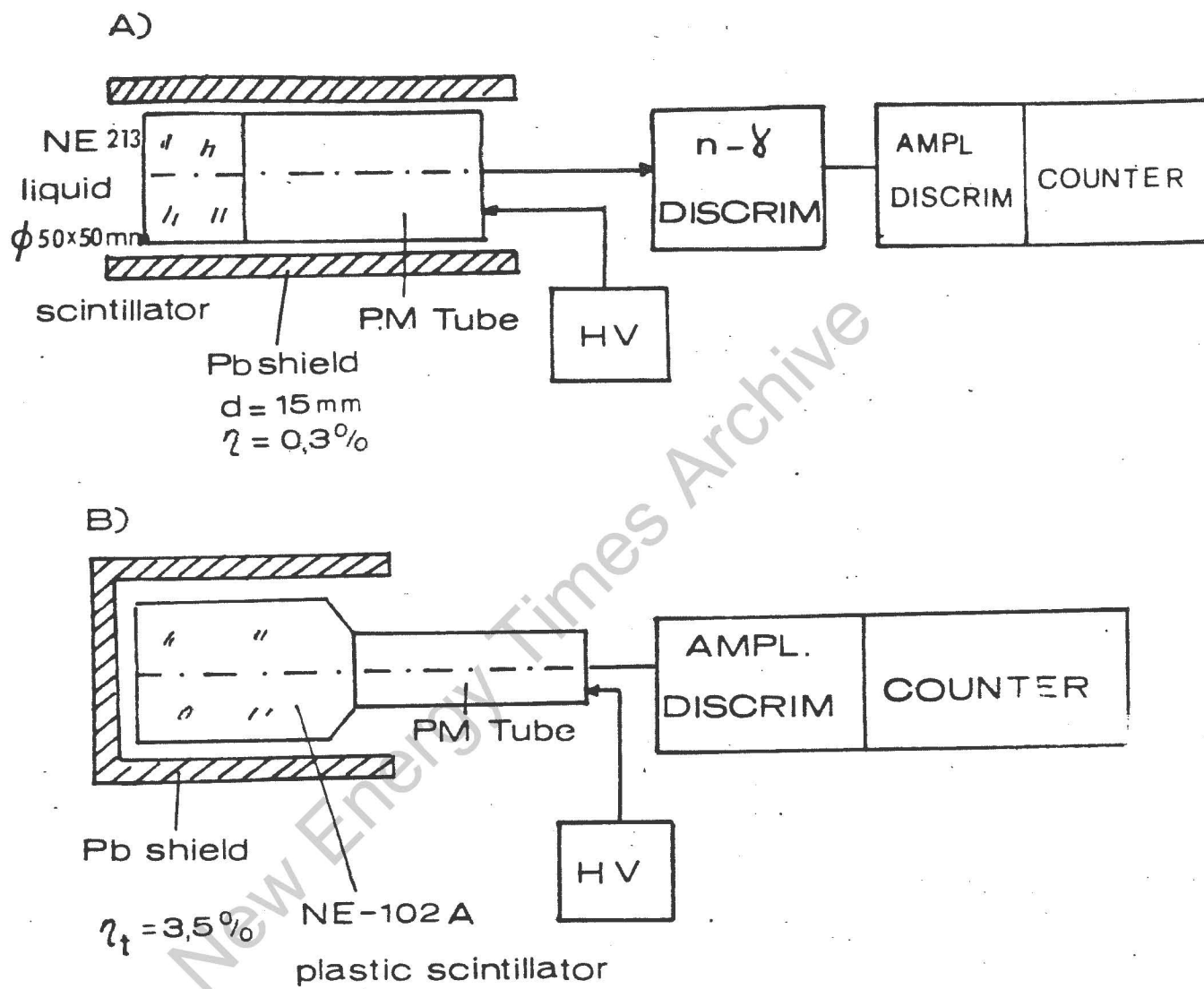
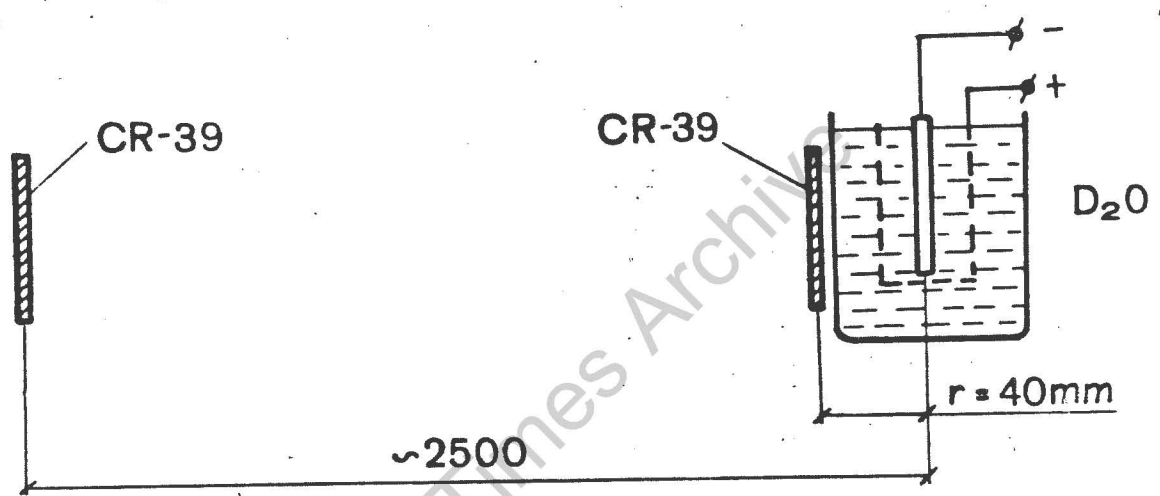


Fig 2

Fig 3



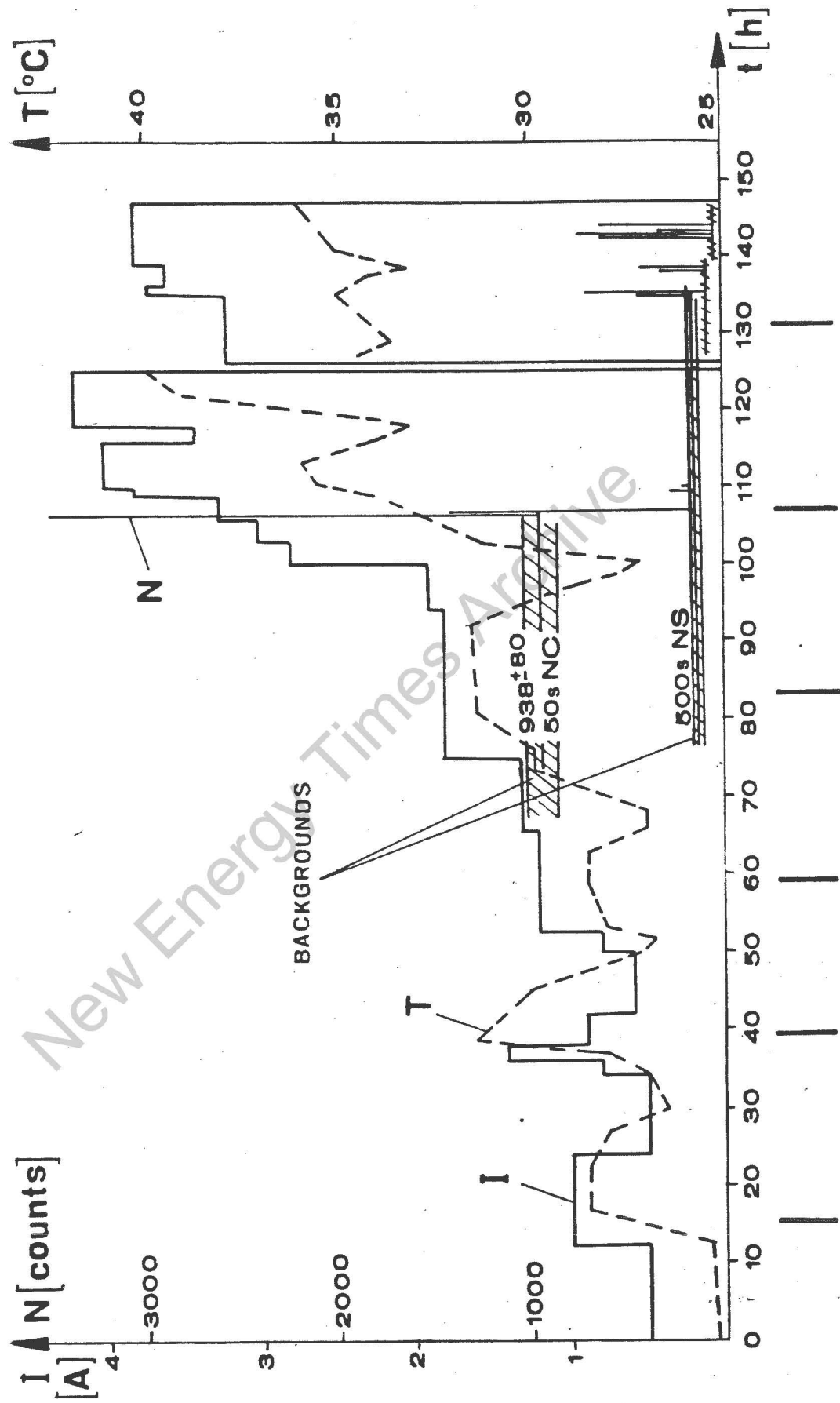


Fig 4

Fig 5

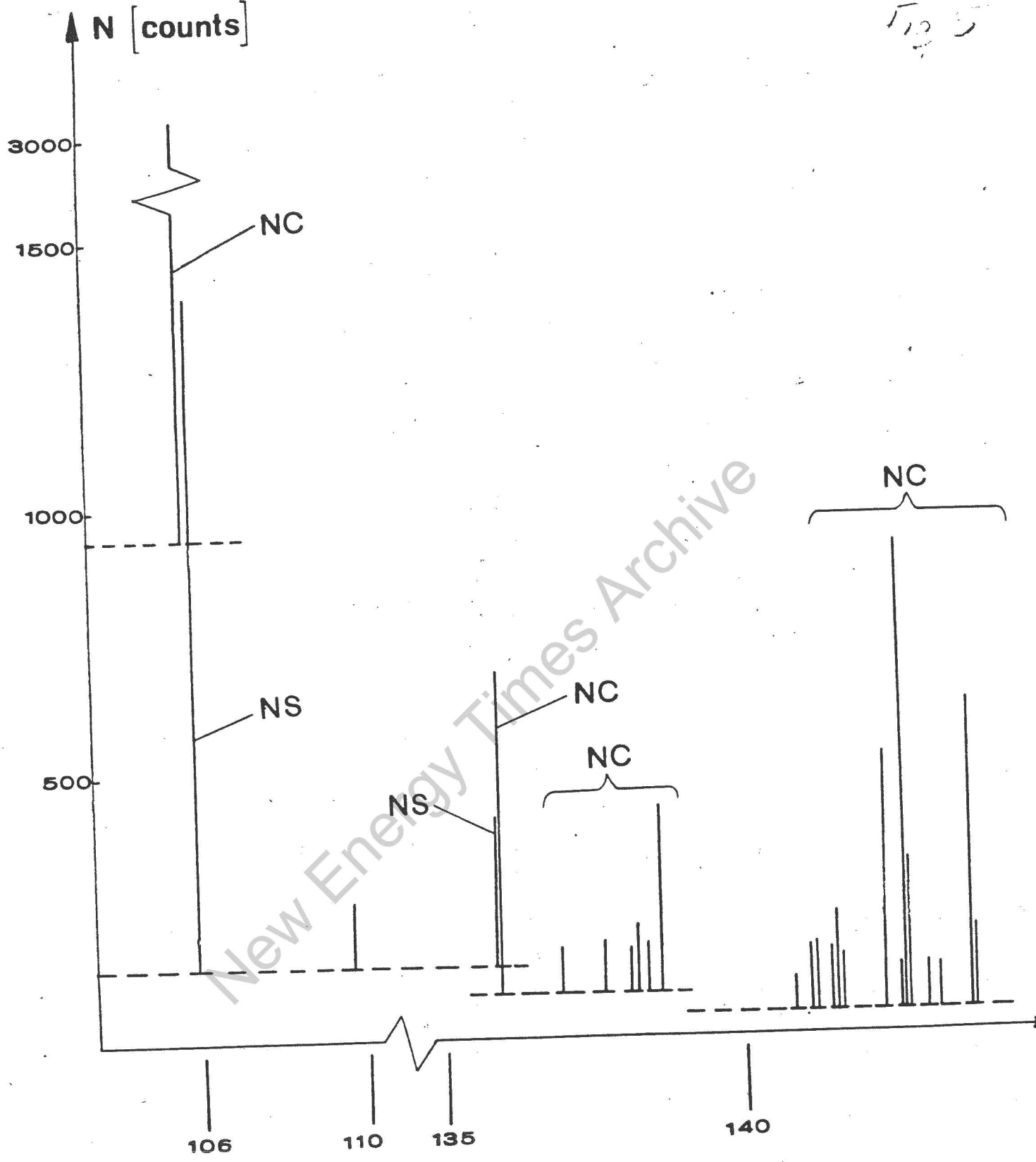


Fig 5